The University of Manchester
Dalton Nuclear Institute

# Managing the UK plutonium stockpile:

# **no easy choices**

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# Foreword

## The UK is considered a world leader in nuclear with a well-established nuclear industry since the 1950s.

This offers many positives for the UK by contributing to current needs in improved energy security and delivering Net Zero, but does present challenges in regards to nuclear legacy management. One of its legacies is its current stockpile of some 140 tonnes of civil plutonium, which currently resides at the Sellafield site in Cumbria. The stockpile originates from reprocessing spent fuel from the UK's reactor fleets, plus some material derived from outside the UK.

The earliest and most widely understood application of plutonium is its use in weapons, however almost all of the UK's stockpile was produced only for civil purposes. In fact, the UK's civil plutonium stockpile is the largest in the world.

So what is "civil" plutonium good for? It could be used as fuel for existing or future thermal reactors. It could also be combined with the UK's 100,000 tonne supply of Depleted, Natural and Low-Enriched Uranium (DNLEU) to fuel new fast reactors, which has the potential to power the UK for centuries. Both options could lead to the reduction of the UK's nuclear legacy burden.

While this all sounds promising, successfully delivering such outcomes would take time, money, organisation, and commitment. Currently, the Nuclear Decommissioning Authority (NDA) is in the process of repackaging the plutonium stocks into more robust containment. Being wary of the current global political and economic climate, it may be that extracting the energy from UK plutonium in the not-too-distant future becomes unnecessarily expensive and political barriers may be too difficult to overcome. Therefore, it might be simpler and cheaper to consider it a waste material alongside the other legacies from the nuclear industry, and safely dispose of it.

So there are some decisions to be made. Some need to be made soon, whereas others can, and should, be safely postponed until we have the necessary supporting information. Delaying a decision does not remove the responsibility associated with it. A concerted and immediate effort to fill these information gaps is needed to put the UK in the best position possible to make the right decision on the right timescale. For this reason, members of the Dalton Nuclear Institute have produced this paper to assist decision-makers in this matter. No final decisions are offered, but instead the paper aims to help drive a conversation to help ensure that the best decisions are made when they are required.

To paraphrase the common saying about building new nuclear reactors, the best time to begin such conversations is around twenty years ago – but the second-best time is now!

**Professor Clint Sharrad**
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# Executive Summary and Recommendations

Over the last six decades, the UK has built up a stockpile of some 140 tonnes of civil plutonium, currently stored as plutonium dioxide powder in Sellafield. The facilities, packaging and conditions of this storage are currently the subject of an NDA improvement programme over the next several decades. After the period of storage, the end point of the plutonium is being examined against the possible futures of conversion into either fuel for nuclear reactors or a wasteform for disposal in the planned UK Geological Disposal Facility (GDF).

The optimal solid forms for disposal and for use as future fuel are distinct, so converting the stored material for delivery of one option will foreclose alternatives. It is therefore advisable to reach a decision on the ultimate endpoint before taking firm steps in any direction.

As nuclear fuel, the plutonium would generate quantities of low-carbon energy, the amount of which would vary with the reactors and fuel types used but could be very significant for the UK. As waste, no energy would be generated, but this option could be expected to reach an earlier end point than use as fuel and to involve less initial cost as part of the overall GDF programme.

To illustrate the benefits and detriments of these futures, this paper examines the basic attributes of, and activities involved with, plutonium storage, use as fuel, and disposal as waste. It does this in broad terms in the body of the paper, with a greater level of background and detail in appendices. The factors revealed by this examination gave rise to the inclusion of "no easy choices" in the study's title.

The key initial finding is that the current programme of improvement at Sellafield is essential to reduce the risks and hazards of plutonium storage, and this must be a priority for resources and funding over the next several decades. This unavoidably long timescale allows time for the choices between "Use" and "Disposal" to be properly examined: essential when any choice will almost certainly exclude any later move to another choice. Time is, however, a consideration, when the storage of plutonium dioxide powder is more hazardous than storing the same plutonium as a reactor fuel or as a wasteform. This will need to be clearly factored into decision making, especially when the desire to minimise hazard/risk appears to be taken as an absolute driver in some quarters.

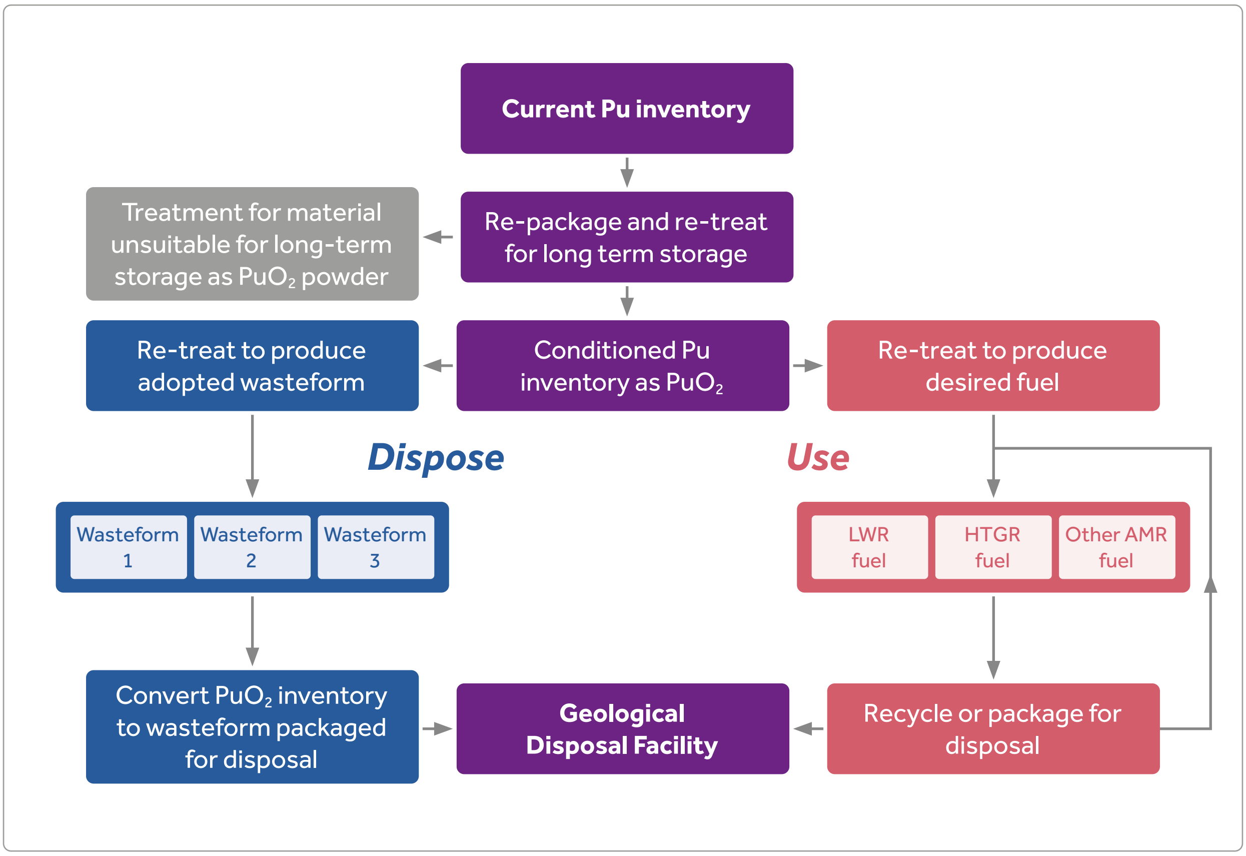

**Figure 1. Plutonium inventory flowsheet choices.**

Overall, the message from this study is that the current storage improvement programme is an essential first step. This will allow the time for a properly resourced process to examine the ultimate fate of the plutonium stockpile on the basis of a balanced assessment of all aspects of the two alternative futures, the pros and cons of which are examined at length in this paper and its appendices. This evaluation is a responsibility of Government. "Decide in haste, repent at leisure" has been a long-time feature of UK decision making in nuclear power – and this is unavoidably an underlying theme for much of this study. It is inevitable that the varied drivers represented within Government will complicate the choice of the way forward; Security, the drive to Net Zero, and the view of the Treasury are typical examples. The interactions of these national policies make this a government-level decision. However, the decision timescales presented in the study allow ample time for a methodical and open decision-making process.

Ten recommendations are presented throughout the paper, in addition to a final, overarching recommendation which summarises the global issue and offers high-level advice for its resolution:

### *Recommendation one*

***Before attempting to make and implement policy decisions regarding plutonium, Government should ensure that a national dialogue takes place allowing stakeholders from all sides to share their views and participate in a respectful, evidence-based debate. This must be more than either a "lip service" consultation with outcomes already decided or a polarised "black versus white" argument. It needs to be an open dialogue, facilitated and led by trusted voices and based on a clear view of Government's thinking of the role (if any) plutonium might play in meeting future UK energy needs.***

### *Recommendation two*

***The current programme of repackaging and storing the plutonium inventory in optimal conditions must be carried out by the NDA and Sellafield Ltd to the currently programmed end point of 100-year design life storage. This provides sufficient time for the necessary comprehensive Research, Development and Innovation (RD&I) required to underpin the whole plutonium lifecycle to be carried out in parallel.***

### *Recommendation three*

*Bringing the UK plutonium programme to a successful end will take several decades at least. Both Government and NDA should provide the commitment and resources needed to ensure continuity and development of capability over this timescale.*

### *Recommendation four*

*Government should decide on and implement a preferred end point for the plutonium once a satisfactory assessment of options and their attributes is available, taking into account changes in storage environment and the hazard that plutonium presents.*

### *Recommendation five*

*Given the hazard represented by the plutonium stockpile and the long duration of plutonium storage, the storage infrastructure is critical to safety and security, so Government, NDA and other stakeholders must ensure that sufficient attention and resources are devoted to long term care of these assets.*

### *Recommendation six*

*The hazard represented by the plutonium stockpile would be greatly decreased by conversion from dispersible powder into a solid form, but the choice of form will determine which future option is to be followed. Government should ensure that a comprehensive assessment is carried out on the attributes and costs of the range of options.*

### *Recommendation seven*

*The different disposition options follow very different pathways to putting plutonium beyond reach. To underpin decision-making, Government needs to develop a full understanding of the whole plutonium lifecycle for each pathway before committing to irrevocable decisions.*

### *Recommendation eight*

*Because of the major uncertainties associated with the UK's plutonium management programme, it is unwise to rely on discounted costs to evaluate the programme and, in particular the assumption of cost decrease associated with discounting should not be used as a pretext to delay decision making and action.*

### *Recommendation nine*

*The decades-long, highly challenging programme needed to address the challenges of the UK plutonium stockpile can only be delivered by an experienced community of practice so Government should ensure there is a sufficient supply of suitably qualified and experienced personnel to deliver the programme.*

### *Recommendation ten*

*Government should ensure that a robust, long-term programme of Research, Development and Innovation (RD&I) is in place to support selection and implementation of any plutonium management option.*

### *Overarching Recommendation*

*Government, which is ultimately responsible for management of the UK's plutonium stockpile, should acknowledge that this is an unavoidably complex, multi-generational undertaking, requiring ongoing stewardship prior to an irrevocable decision on the end point for the material, and should put in place suitable arrangements. There are significant major uncertainties which can only be managed through a long term, programmatic approach with continuity, flexibility, adaptability, underpinned by Research, Development and Innovation (RD&I) commensurate with the scale of the challenge. This RD&I will not only decrease uncertainties and provide opportunities to accelerate the programme and reduce cost but will also be essential in developing the specialist community of practice required for delivery. The recently concluded Advanced Fuel Cycle Programme may be a useful model.*

# 1

# Introduction

The UK has a history of electricity generation from nuclear power spanning nearly 70 years, comprising three generations of reactors, all powered with uranium-based fuels. Standard operation of nuclear power reactors produces plutonium from $^{238}U$ (uranium-238, the isotope which makes up the large majority of uranium fuel) by neutron capture. Much of the UK's spent fuel was reprocessed after use to recover useful material, with the plutonium separated for future use, mainly with the intention of eventually utilising it to start generation using a future programme of fast reactors. As will also be examined, the economic and resource drivers for the adoption of fast reactors did not materialise, and the UK's plutonium inventory remains stored. The Climate Change Act 2008 specifies that [1, Sec. 1]:

> *"It is the duty of the Secretary of State to ensure that the net UK carbon account for the year 2050 is at least 100% [amended from 80% in 2019] lower than the 1990 baseline."*

The 2019 commitment to achieving "Net Zero by 2050" has changed the future energy policy of the UK, with low-carbon energy sources a focus for the future as a result. Nuclear energy, while demonstrably very low carbon, still must demonstrate its economic effectiveness and achieve acceptance by a broad range of stakeholders.

Viewed against this background, the UK's plutonium stock could have a significant role in both generating low-carbon energy, and in facilitating the huge energy potential of the UK's stock of depleted uranium (which becomes a viable energy source in fast reactors in partnership with plutonium). Against this, doubts on the economics of plutonium use, together with safety and security fears around its continued storage, can drive options for its earliest possible disposal as waste. The choice is complicated by the fact that the timescales of all the "Use" or "Dispose" options are dependent on construction and operation of new plants, and many also require the availability of the UK's projected Geological Disposal Facility (GDF). These factors mean that none of the *Use or Dispose* options can be completed in fewer than several decades.

This paper attempts a dispassionate analysis of the current situation, and of the various potential futures. It mirrors its title in recognising from the start that there are "No Easy Choices" – all the future paths must overcome significant barriers and have the potential to appear sub-optimal in hindsight. The objective here is to provide a similar level of clarity for each choice – equally encompassing the good, the bad, and the ugly.

The position, both in the literature and in the stakeholder consciousness, is complicated by the fact that plutonium has in many circles, been "the element that dare not speak its name". The concepts, arguments, and data in the field of mapping plutonium choices are unavoidably complex, but this paper seeks to be accessible to the mainstream reader. The approach has therefore been taken to pitch the main paper in plain English, without complicating the text with multiple footnotes and references. This would however leave many questions unanswered, so detailed appendices have been compiled which contain the technical content, with references to supporting material.

## 1.1 Public, Policy and Societal Context

Plutonium holds a special place in the public consciousness. This is probably largely due to the association with weapons, the fact it is perceived to be a deadly hazard to humans and the notion that it is an artificially produced element, not found in nature.

These beliefs are only partially true. Plutonium does occur in nature – albeit in trace quantities, and it is the element with the highest atomic number to do so. Natural plutonium is produced in $^{238}$U deposits when one $^{238}$U atom captures neutrons emitted by the decay of another.

And while plutonium is hazardous, as primarily an alpha emitter it does not present the same hazard as the most highly radioactive wastes that require massive shielding. In fact, relatively lightweight glovebox protection is all that is required. The risk to health is much higher if plutonium-containing material is inhaled through mouth or nose, or if the material enters the body through a cut or scratch.

The weapons connection dates to the Manhattan Project. The "Gadget", detonated during the Trinity test in July 1945, and the Nagasaki bomb of August 1945 had plutonium cores, for which the material had to be produced in significant quantities for the first time. It was first synthetically isolated in 1940 at the University of California, but the news was embargoed until 1948 due to wartime secrecy concerns.

Scholarly papers about plutonium and its public perception have been written since the 1970s [2]. Even today, those who work in the nuclear sector are sometimes wary using the word "plutonium" to describe the work they do, choosing instead to talk of "special nuclear materials" or "alpha materials". The UK's current Alpha Resilience and Capability (ARC) Programme is a prime example, describing its vision as [3, p. 2]:

> *"To sustain and enhance our world leading Alpha capabilities so the UK nuclear industry is ready to enact critical national Programmes."*

It seems likely that the heightened public sensitivity to considerations of plutonium – irrespective of the quantities involved, or the nature of the discussion – acts as an obstacle to both rational and open discussion of issues surrounding its use and any kind of policy decisions on potential strategies for management or re-use.

Policymakers have a choice. They can simply accept this, recognise the risk, do nothing, and hope for the best; or they can take steps in advance (ideally a long time in advance) to make sure that responses to any such announcement are more objective, rational, and fact-based.

That requires a discussion, perhaps even a communication campaign, where the challenges of dealing with plutonium are discussed along with the issues around its creation, management, and disposal. This must be done in a pragmatic and respectful way, with strong recourse to evidence and with efforts made to identify and remove misconceptions on all sides. The rights of participants to be concerned must not be diminished.

To increase the chances of such discussions being constructive and respectful, it is important to give all parties the chance to air their concerns and have their questions answered. It is also vital to identify and involve people (expert or otherwise) who will be regarded as trusted voices by as many people as possible.

Previous examples of stakeholder engagement around such thorny issues [4] have demonstrated that it is possible to reach either a (sometimes grudging) agreement or else a more mutually respectful disagreement on key issues. But such measures take time and commitment from all concerned.

Identification of trusted voices is key – but there are intermediaries such as the Science Media Centre or the Parliamentary Office of Science and Technology who can ensure that technical specialists have the right combination of specific knowledge and interpersonal empathy to play a part in such conversations. But the floor can never be solely occupied by scientists and other "experts". It is important to hear from environmental activists, community groups, regulators, media, employees (current or potential) and labour unions as well.

***Recommendation one:***

***Before attempting to make and implement policy decisions regarding plutonium, Government should ensure that a national dialogue takes place allowing stakeholders from all sides to share their views and participate in a respectful, evidence-based debate. This must be more than either a "lip service" consultation with outcomes already decided or a polarised "black versus white" argument. It needs to be an open dialogue, facilitated and led by trusted voices and based on a clear view of Government's thinking of the role (if any) plutonium might play in meeting future UK energy needs.***

# 2

# The Plutonium Stockpile

This section summarises basic information on the origins and current state of the UK plutonium inventory. It covers the origins and size of the stockpile and the factors which have led, over the decades, to the current position where around 140 t* of plutonium is stored, mainly as plutonium dioxide ($PuO_2$) powder, and almost all at the Sellafield site. A fuller explanation with multiple references is given in Appendix 1, which should be the first port of call for queries concerning content in this chapter.

The UK started manufacturing plutonium for military purposes in 1950, reprocessing the fuel from the Windscale air-cooled, graphite-moderated piles. After closure of the Windscale Piles in 1957, reprocessing continued with spent fuel from the dual purpose (i.e. military plutonium production and electricity generation) Magnox reactors at Calder Hall and Chapelcross. The magnesium-clad, uranium metal fuel of these reactors was designed for reprocessing and storage of the spent fuel for long timescales was problematic. A series of civil (i.e. not for military application) Magnox reactors followed, and reprocessing was continued in order to provide the initial fuel for a series of future fast reactors. Fast reactors were presumed to be necessary in the future because of an anticipated imminent† world shortage of uranium due to large scale nuclear power rollout worldwide. In reality, world nuclear programmes did not increase as quickly as had been assumed, and it became apparent that availability of uranium resources had been much underestimated. However, because of the nature of the fuel, reprocessing was obliged to continue until the end of the Magnox programme, with the last reactor ceasing generation in 2015 and Magnox reprocessing ending in 2022 [5, p. 6].

Fuel from the second generation of reactors, the civil Advanced Gas-cooled Reactors (AGRs), was not obliged to be reprocessed, though some (but by no means all) fuel was reprocessed for commercial reasons. This programme coincided with the construction of the THermal Oxide Reprocessing Plant (THORP, commissioned in 1994), whose principal mission was to reprocess fuel from the UK and also from foreign reactors (for which charges were levied). This activity has led to some foreign-owned plutonium remaining in the UK, with the position detailed in Appendix 1.

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** **t** denotes metric tonne, equivalent to 1,000 kg. A tonne is not equal to and should not be confused with either the short ton or long ton, which may appear in US or historic literature. t is used here throughout and may appear as tHM (tonnes of heavy metal), often used to quantify materials such as uranium, plutonium and their derivatives.*

*† **Imminent** in reactor delivery terms, i.e. of the order of tens of years.*

By this time it was accepted that the economic drivers for a fast reactor programme had not materialised, and the UK had started to accumulate a significant stockpile of plutonium which would eventually grow to around 140 t, as shown in Table 1.

**Table 1. Origin of the UK's plutonium stock. See Appendix 2 for more detail.**

| Origin | Tonnes of material |
|---|---|
| Magnox | 85.8 |
| AGR | 23.6 |
| Foreign plutonium transferred to the UK | 8.3 |
| Foreign owned | 24.1 |
| **Total** | **141.8** |

A complication with quantifying plutonium is that it varies in its nuclide composition depending on the parent fuel's burnup and irradiation conditions, and also in its physical properties such as particle size and purity. Variations in both these categories can affect the future treatment routes of the material. As discussed in Appendix 1, the condition of some of the $PuO_2$ and its storage containers has deteriorated with time, leading to the need to repackage the material, and examine the potential need for treatment or disposal of the handled material. It has also been acknowledged that, as the material must be stored for at least another few decades, more durable arrangements are essential, and Sellafield has embarked on a programme of repackaging and committing the material to a more robust storage regime with a hundred-year design life.

***Recommendation two:***

***The current programme of repackaging and storing the plutonium inventory in optimal conditions must be carried out by the NDA and Sellafield Ltd to the currently programmed end point of 100-year design life storage. This provides sufficient time for the necessary comprehensive Research, Development and Innovation (RD&I) required to underpin the whole plutonium lifecycle to be carried out in parallel.***

# 3

# Ways Forward – Choices for Now and the Future

The context of this section is explored in some detail in Appendix 2, which also contains a body of references to published material in the field.

## 3.1 Government Policy

In 2016, Andrea Leadsom, the minister for the Department of Energy and Climate Change (DECC) stated [6]:

> *"Over the past decade the Government, supported by the Nuclear Decommissioning Authority (NDA), have developed the policy and strategy framework for managing the UK's inventory of separated civil plutonium. We are working closely with the NDA to ensure the safe and secure storage of the material at Sellafield and to plan, develop and implement a management solution for separated civil plutonium in the UK until the inventory has been reduced to zero and is put beyond reach. A key focus of that strategy is hazard reduction, which means addressing Sellafield's legacy facilities. Putting the material beyond use will take many decades, so we therefore need to ensure that all nuclear materials are stored in modern facilities that are safe and secure."*

Adding:

> *"...a decision on how to proceed cannot and will not be taken quickly. It is about making the right decision at the right time, underpinned by the right evidence. It is important to note that any decision will take many decades to implement, which is why a decision on plutonium disposition should not be made in isolation. There are interdependencies across the new nuclear build programme, geological disposal and national security outcomes."*

For the decades that the plutonium inventory has been in existence, it has been recognised that, ultimately, the storage period must be finite, and must be followed by options to either use the plutonium as nuclear fuel, or to dispose of it in a suitable wasteform in the UK's GDF, when this becomes available. In 2011, re-use was the preferred route [7, p. 26]:

> *"The UK Government has concluded that for nuclear security reasons the preferred policy for managing the vast majority of UK civil separated plutonium is reuse and it therefore should be converted to [mixed oxide] fuel for use in civil nuclear reactors."*

However, it was recognised that there was nowhere near enough knowledge available to make a final decision. Andrea Leadsom, again in her 2016 statement to the House of Commons, summed-up the position in some detail, concluding that [6]:

> *"The Government remain open to any credible option for managing the inventory, but of course it must offer the best value for money to the taxpayer. Only when the Government are confident that our preferred option could be implemented safely and securely with an eye to cost – that it is affordable and deliverable and offers value for money – will we be in a position to proceed."*

This statement also mentioned that much work was already proceeding to improve the existing storage regime, and a current programme is underway to repackage the $PuO_2$ inventory and place the new containers in a refurbished store with a design life of 100 years. This is further examined within the options discussed below.

***Recommendation three:***

***Bringing the UK plutonium programme to a successful end will take several decades at least. Both Government and NDA should provide the commitment and resources needed to ensure continuity and development of capability over this timescale.***

## 3.2 **Driving Decisions – Weighing Options**

Broadly, the future fate of the plutonium inventory comprises three choices, which will be discussed in turn:

1. *Status quo* – continued storage and its developments, though it is recognised that storage cannot be indefinite.
2. *Use* – methods of using the inventory as fuel in reactors to generate low-carbon energy.
3. *Dispose* – disposal of the plutonium, with several wasteforms and disposal methods currently being studied.

The decision between options for the future fate of the UK's plutonium inventory could clearly involve, inter alia, safety, environmental and security risks, the emission or the prevention from emission of large volumes of carbon, and notably large sums of money. In an ideal world, these could all be reduced to a common unit, with the ideal answer being "the option that gives the best overall result". In the current reality such a world does not exist, and the ideal decision-making process must move in the direction of "finding the best solution with a clear explanation for why and how the choice has been made".

Fortunately, there is enough knowledge and experience to plot a credible (but not necessarily accurate) view of the range of activities required for *Status quo*, *Use* or *Disposal* of the inventory. This will involve broad estimates of the plants to be built, timescales of operation and costs, which will allow at least a credible view of the various futures and their carbon impact. However, no matter how accurate, these estimates will not allow for a decision without weightings being placed on the individual parameters – for example: "the overall preferred option is Option X, because although it costs more, we believe that the reduction in security risks dominates the answer".

It is inevitable that the varied drivers represented within Government will complicate the choice of the way forward. "Security", the drive to "Net Zero", and "the view of the Treasury" are typical examples which will mean that, even when all the data is known, the "weights" given to the various factors will vary from department to department. However, the decision timescales presented in the study allow ample time for a methodical and open decision-making process.

One clear factor to be evaluated is that even early steps along some future paths can effectively preclude moving to another path if situations change. History clearly illustrates this as the UK's plutonium strategy was designed under the assumption that uranium would rapidly become scarce and expensive – which did not happen. Similar assumptions in future paths must be explicitly defined. Another complicating factor is that minimising hazard/risk is taken as an absolute driver in some quarters – which would immediately drive towards converting the stored $PuO_2$ powder into a solid form. This would almost certainly rule out the option of using the material to generate energy, and the current NDA programme to optimise the $PuO_2$ powder and its storage is considered adequate to remove the need for short-term conversion to solid.

Appendix 4 gives more detail of the scientific background of the current material status and key knowledge gaps associated with ongoing interim storage, as well as manufacture into fuel or a disposal wasteform.

***Recommendation four:***

***Government should decide on and implement a preferred end point for the plutonium once a satisfactory assessment of options and their attributes is available, taking into account changes in storage environment and the hazard that plutonium presents.***

## 3.3 **Continued Storage**

In view of the programme to repackage the plutonium inventory, and the inevitable time it will take to design and build plants to pursue the alternative programmes examined in the following sections, storage will remain a fundamental element in future programmes for at least some decades to come.

The key factors which need to be considered for the *Status quo* option are the form of the plutonium and the specification and condition of the packaging and stores. Provided that all these elements are maintained in a good condition, continued storage will continue to be considered "safe" (i.e. that the risks of losing containment and suffering a plutonium leakage are considered adequately low). This infers that the current plans, which, as will be seen, all involve storage for several decades, can be regarded as safe – while relying on maintenance and operational safety being maintained at the necessarily high level.

The Royal Society [8] considered the hazard of storing powder was high enough to recommend converting separated plutonium into Mixed OXide (MOX) fuel as soon as it was feasible to do so. This was based on the assumption that suitable reactors could be identified in advance so that the MOX fuel manufactured matched the fuel specification. This reactor-specification-matching seems problematic with the currently ill-defined reactor build programme – and in any case the phasing of the MOX fuel plant will require study, as discussed in the *Use* scenarios in Section 3.4.

***Recommendation five:***

***Given the hazard represented by the plutonium stockpile and the long duration of plutonium storage, the storage infrastructure is critical to safety and security, so Government, NDA and other stakeholders must ensure that sufficient attention and resources are devoted to long term care of these assets.***

***Recommendation six:***

***The hazard represented by the plutonium stockpile would be greatly decreased by conversion from dispersible powder into a solid form, but the choice of form will determine which future option is to be followed. Government should ensure that a comprehensive assessment is carried out on the attributes and costs of the range of options.***

It should, however, be noted that the potential detriments from episodes involving malicious intent (see Section 3.6) will remain at a comparatively high level for at least as long as the $PuO_2$ remains as a fine powder in relatively transportable containers. The level of precaution implemented thus far will clearly need to be maintained at this appropriate level as long as $PuO_2$ powder storage continues.

## 3.4 **Use as Fuel**

As already discussed, the original driver for generating a UK plutonium inventory was to fuel an expanding fleet of fast reactors in response to impending uranium shortages. When these shortages did not occur, the growing inventory was, and remains, stored. The option of burning it in reactors is, however, still available, and this section examines the use of the plutonium as fuel in future reactors in the UK.

The reactors making up the current Government target of 24 GWe of nuclear generation by 2050 are assumed to be a combination of Light Water Reactors (LWRs) and Small Modular Reactors (SMRs)*. A fleet of High Temperature Gas-cooled Reactors (HTGRs) is also being discussed, principally for high temperature heat provision, but the fleet size has not been quantified.

The 140 t plutonium inventory would convert into around 2,000 tHM of 7% [9, p. 11] plutonium Pressurised Water Reactor (PWR) fuel. Each year a LWR loads around 17 tHM of fuel per GWe, and for reactors using MOX, a third of this (5.67 tHM) would be MOX. Therefore, MOX loading will last around 350 reactor-years for a 1 GWe reactor – or the equivalent of around ten 1 GWe reactors for 35 years.

The amount of power generated from the MOX fuel at a typical burnup of 55 GWd/tHM would be around 107 gigawatt-years of electricity – this is equivalent to just around four and a half years of total output of the 24 GWe fleet, or nearly three times the UK's total electricity generation in 2022†. These figures are illustrative to give a feel for the potential for the useful energy that could be extracted from the plutonium if used in MOX fuel on a second cycle basis.

Neither of the reactors being built (i.e. the two at Hinkley Point C) or those which appear imminent (two planned at Sizewell C) will be constructed to receive plutonium MOX fuel, though they could be modified to do so. The reactor at Sizewell B was built as MOX-ready but the relevant features were subsequently removed during maintenance.

The position with the PWRs is thus that the projected UK programme could accommodate the plutonium inventory as MOX fuel, but the reactors would need to be enabled to accommodate it, and a successful outcome achieved for the economics of manufacture and use of the fuel.

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* *To maintain consistency with previous papers, we adopt the following nomenclature concerning reactor systems: GW-sized PWRs, such as the EPR at Hinkley Point C, are referred to as LWRs; the anticipated SMRs are essentially smaller versions of LWRs; with any other advanced reactors such as HTGRs considered Advanced Modular Reactors (AMRs).*

† *A commonly shared rule of thumb is that an LWR loads ~30 tHM of fuel per year, per GW. This is broadly correct for a reactor such as Sizewell B with an average (original) enrichment of 2.6% $^{235}U$, and an average burnup of ~33 GWd/tHM. Enrichment for a reactor such as those being built at Hinkley Point C is typically 5.5% $^{235}U$, with a higher burnup of ~55 GWd/tHM. Assuming a load factor of 90% and net efficiency of 35.4%, the average fuel loading per GWe is: [0.9 × 365 / (55 × 0.354)] = 16.9 tHM/year.*

The spent MOX fuel would have a plutonium content of around 5%. This would be suitable for disposal or could be reprocessed to power advanced reactors. Of course, the plutonium inventory could be used on a longer timescale to act as the "nucleus" of a fast reactor fleet. While timescales and fleet size are too conjectural to make meaningful predictions, it is significant that the 140 t UK plutonium inventory could start up a fleet of around 6-10 GWe of typical fast reactor technologies, assuming three fuel loadings of 5-7.5 t of plutonium are required for each GWe, after which point their closed fuel cycle becomes self-sustaining. It is also noteworthy that the $^{241}$Pu component of the plutonium will be lost by decay during prolonged storage, as the half-life of $^{241}$Pu is 14.3 years. As $^{241}$Pu is fissile this will reduce the fissile fraction of the plutonium, which is mainly of concern to the application in thermal reactors. The $^{241}$Am daughter product of $^{241}$Pu does not significantly contribute to the fissile content of fuel, but does present some handling issues. Apart from $^{238}$Pu which is a minor constituent, the half-lives of other plutonium isotopes are too long to become a storage issue.

The manufacture of PWR MOX fuel would require a new plant, as the abandoned Sellafield MOX Plant (SMP) is thought unlikely to be worth resuscitating. From the overall capacities discussed above, an annual production capacity of around 60 tHM/y would serve a 30-year MOX burning programme in around 10 reactors.

There is a choice of industrially proven technologies between the Short Binderless Route (SBR) as used in SMP, or the MIcronized – MASter blend (MIMAS) route used in the French programme. Both have been proven to produce satisfactory fuel, and though the SMP was viewed as a failure, this was due to poor engineering and plant design rather than any deficiencies in the fuel route. Recent studies have generally concentrated on the use of the MIMAS route. Note that, as discussed previously, there would be the possibility of manufacturing the MOX fuel pellets as soon as their specification was known and the MOX plant was available. This would need to be analysed nearer the time, but it seems unlikely to represent a large reduction in storage time in powder form.

It should, of course, be noted that the manufacture of MOX pellets will inevitably lead to some being out of specification, and there is also the likelihood of out-of-specification batches of $PuO_2$ powder. This will necessitate a method of recycling this material, with many challenges likely to be amenable to a combination of sintering with a hydrogen-containing atmosphere, teamed with oxidation-reduction (OXRO) cycling.

This analysis has not covered the possibility of using plutonium in any HTGR programme. This would of course be possible, but would require a new plutonium MOX TRISO fuel* plant, and the timescale at which an adequate reactor programme might be available is currently long-term and uncertain.

## 3.5 **Disposal**

Although surface stores for radioactive waste can plausibly have a life of around 100 years, the UK's strategy for permanent disposition of High and Intermediate Level Waste (HLW and ILW, respectively) is disposal in a GDF. This is waste that has accumulated mainly from nuclear power generation but also smaller amounts from other medical, industrial, research and defence activities. A GDF will be a highly engineered structure, located underground from 200 to 1,000 m deep and with a footprint of 10-20 km$^2$ [10, p. 9]. The GDF will present multiple barriers, both engineered and natural, to radionuclide migration, ensuring that the risk to the biosphere from geological disposal is acceptably low. Currently, several candidate host communities are under consideration and some early site investigations have been carried out. Current timelines estimate that a GDF could be ready to begin receiving ILW by ~2060 and HLW and spent nuclear fuel from 2075, with filling and close out running into the next century.

With the current plutonium retreatment and repackaging project at Sellafield due to complete in ~2060 this would present two options: manufacture the wasteform as soon as possible, with interim storage until the GDF is able to receive plutonium-bearing wasteforms; or begin manufacture of the wasteform to match the ability of the GDF to receive it. Ultimately, unless it is used in a closed cycle fast reactor programme, the plutonium stockpile is destined for a GDF, whether it be directly as a disposal wasteform or indirectly as spent MOX fuel. In total, it is estimated that this plutonium-bearing waste will generate ~12,500 m$^3$ of packaged waste volume in either scenario. With a total waste package inventory of ~773,000 m$^3$ [11, p. 17], this would account for less than 2% of the GDF inventory by volume.

Disposal wasteforms (discussed further in Appendix 4) for $PuO_2$ have typically been based on naturally occurring ceramic minerals which contain uranium and thorium and that are known to be stable over geological timescales. This has led to the downselection of single-phase pyrochlore and zirconolite ceramics as the main candidates for immobilisation of the plutonium and disposal in the GDF. Neutron absorbers such as gadolinium and hafnium may well be incorporated to prevent post-closure criticality. However, such materials have not been manufactured at scale with plutonium. In addition, there is insufficient understanding of their long-term radiation and leaching behaviour to support the safety case for disposal in a GDF.

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** **TRISO fuel** is TRi-structural ISOtropic fuel; fuel particles used in HTGRs and some other reactors. HTGR fuel could also be oxycarbide, not oxide.*

Novel manufacturing processes such as Hot Isostatic Pressing (HIP) may offer benefits for manufacturing of plutonium-bearing zirconolites or pyrochlores. In this method, the powdered feed material is loaded into a specially designed, sealed metallic canister and subjected to high pressure gas at elevated temperature, producing a consolidated canister containing a monolithic sintered product. This has several advantages as it offers increased wasteform stability, volume reduction and reduction of secondary wastes (such as volatile off-gases). However, the technology is much less mature than conventional sintering techniques and would need demonstration at scale prior to a full plant design and operation.

An alternative wasteform is so-called "disposal MOX". This would be similar to fuel MOX but probably manufactured in larger pellets, with appropriate specifications on pellet quality and finish, and potentially incorporating neutron poisons. However, the specifications are yet to be defined for a disposal MOX product. The advantage of disposal MOX as a wasteform is that industrial scale manufacture of plutonium-bearing MOX fuel has been demonstrated internationally and there is a large amount of post-irradiation data for spent fuel to predict radiation damage behaviour during disposal. Disposal MOX manufacture would likely be based on an industrially proven fuel MOX manufacturing route such as SBR or MIMAS. Once a disposal MOX specification was available, trials could be undertaken to demonstrate feasibility and support selection of manufacturing route, leading to plant design and construction ready for disposal MOX manufacture.

***Recommendation seven:***

***The different disposition options follow very different pathways to putting plutonium beyond reach. To underpin decision-making, Government needs to develop a full understanding of the whole plutonium lifecycle for each pathway before committing to irrevocable decisions.***

## 3.6 Benefits, Detriments, Risks and Hazards

As has already been introduced, the possible ways forward have a variety of benefits and detriments in terms of energy generation as an upside, and ongoing risks, hazards and cost as downsides. The cost elements will be examined in Section 3.8, while assessment of the risk and hazard elements, which is rather more difficult to judge and quantify, will be attempted here.

The risks and hazards of stored $PuO_2$ arise from the unplanned release of the stored material leading to doses to the workforce and the public. Releases can be due to failure of plant containment or operational problems (e.g. container failure). Alternatively, "malicious intent" can be involved where containment failure is due to sabotage, or where plutonium is stolen and used for a malicious purpose – for example as material for release as part of a "dirty bomb".

It is inherently difficult to quantify the absolute relative risk/hazard from malicious intent, especially in an open literature document. This is a major barrier to clarity, as malicious intent is the major driver for much of the concern about the plutonium inventory and its storage.

For releases due to plant and operational problems, however, it is possible to assess the relative risk/hazard by considering the state of the stored material (e.g. $PuO_2$ powder generally has a small particle size and spreads easily if released) together with the specification and condition of the storage plants and their operation (for example, the current repackaging and storage programme at Sellafield is being driven by the sub-optimal storage conditions).

The 2011 Royal Society study recommended reduction of hazard by the prompt conversion of $PuO_2$ powder to MOX pellets [8]. The future use of such pellets as fuel is not assured because at present, neither the eventual reactor designs to be used, or the specification for MOX fuel are known. Also, the current programme to improve $PuO_2$ powder packaging and storage is scheduled to last for several decades, which will much reduce the hazard that the stored powders present.

As will be discussed below, a comparison of the future treatment of the $PuO_2$ inventory will require a comparison across these possible futures, with many dilemmas such as:

- After achieving optimum storage conditions (i.e. those pursued by the current NDA programmes), hazards will further reduce only when the material is changed from powder to a less mobile form.
- However, for the immediate future this change would almost certainly rule out the possibility of using the inventory for generating energy in future reactors.
- Any change to a form suitable for disposal will require considerable work to specify a form which can be manufactured effectively in suitably constructed plants, and also meets the specification for the yet-to-be-identified UK GDF.

Thus, though the arguments for prompt action are significant, arguments for waiting for matters to clarify also have much weight.

## 3.7 SED Methodology

To attempt to assess the risks and hazards from operations with radioactive species, the Safety and Environment Detriment (SED) methodology was developed for NDA by a multi-stakeholder group set up by the Liabilities Management Unit (the precursor of NDA) in early 2004.

Their Prioritisation Working Group [4, p. 23], was tasked to develop and agree a common framework for prioritising the work to be carried out by the NDA's contractors. The aim was to develop a methodology to ensure that the most important things are done first. A key part of the work was the development of measures of hazard potential of wastes, known as the Radiological Hazard Potential (RHP) and the Chemical Hazard Potential (CHP). These quantities were then used to assess the relative potential detriment that materials could cause when stored in defined conditions. The relative potential detriment was termed the SED.

Though the SED result is not a direct measure of hazard, it is driven by well-defined parameters relating to:

- The relative ability of the stored substance (in this case plutonium) to cause doses to people,
- The relative ease of the material spreading into the workplace/surroundings if it were released,
- The suitability of the plutonium store, and
- The nature and condition of the plutonium.

While emphasising that a SED score is NOT an absolutely justified quantity, the formula was put together so that it produced results that seemed sensible after a rigorous examination by a stakeholder team with very broad representation (i.e. industry representatives, regulators, local nuclear site stakeholders and nuclear sceptics). The Prioritisation Working Group all agreed, after examining many examples, that SED gave a sensible "bigger = worse" relative score.

Disposal in a GDF was not considered in the SED methodology development but an element of the methodology assesses how stable things are and how often arrangements need to be checked. As the disposed waste is committed to a repository meeting the GDF disposal safety case, there is no intention to examine or re-treat it, essentially for infinite time. When infinite time is applied to the SED score, that score becomes zero (i.e. the waste is removed from any control regime).

Noting that SED was not developed to cover the risks and consequences of any malicious attempt to cause environmental and human health damage by stealing and releasing nuclear materials (dirty bombs) or by seeking to disrupt/breach the materials' storage, it should be noted that some of the parameters that make up SED, in fact, also shed some likelihood on the relative consequences of such a disruption/breach.

The SED methodology is discussed in more detail in Appendix 3, including malicious releases.

## 3.8 **Economics**

All the possible future paths examined have very considerable costs, which will be commented on where possible. The only paths which may offer some mitigation of the costs are the *Use* options, where an income from energy sales will be generated. As well as the monetary benefit from the generated energy, there should also be the consideration of the benefit arising from reductions in carbon emissions. For *Use* options, the low-carbon credentials of nuclear energy are well referenced and could be further improved by using closed fuel cycles. Care must also be taken that the comparisons between different low-carbon energy sources are accurately assessed. As an example, the usefulness of methods like Carbon Capture, and Storage (CCS) is dependent on the effectiveness of the CCS process, and as such there must be mechanisms for measuring this during operation. CCS with no such monitoring would make a nonsense of any attempts at pursuing a level playing field carbon-reduction strategy. It is also essential that whole lifecycle impacts are considered so that, for example, natural gas leakage during initial extraction is also accounted for. Similarly, the costs and detriments of nuclear energy must also be seen to be holistically taken account of. Initial construction and detriments associated with uranium supply are generally complete, but long-delayed expenditure such as decommissioning and waste disposal, including that in a GDF, must also be included.

Timescales must also be discussed here since the timescales for decommissioning and waste disposal programmes run into decades. A significant reduction in notional programme costs would be achieved if options were being evaluated on the basis of discounted costs on an essentially constant estimate of current cost activities. In reality, however, the activities, methods and regulatory standards have inevitably changed over time. Therefore, the estimates involve activities which are not defined in any detail and take place several decades in the future.

Historically, the costs of nuclear decommissioning have been substantially underestimated. For example the first estimate of the Nuclear Provision (i.e. how much would it cost to clean up the UK's nuclear sites) was £18 billion in 1992, compared with a 2022 estimate of £135.8 billion [12, p. 138]. While the bases on which these two numbers were derived may differ in detail, the estimated cost of decommissioning has clearly risen substantially over the intervening three decades, with much of the increase reflecting greater certainty and better definition of the task. Given the major technical, programmatic and political uncertainties associated with plutonium, it would therefore be very unwise to assume that discounting over the programme's long life will necessarily diminish the real terms costs. As an example, the NDA's 2019/20 accounting reveals total undiscounted liabilities of £131.6 billion, less than the discounted liability of £134.9 billion [13, p. 178].

***Recommendation eight:***

***Because of the major uncertainties associated with the UK's plutonium management programme, it is unwise to rely on discounted costs to evaluate the programme and, in particular the assumption of cost decrease associated with discounting should not be used as a pretext to delay decision making and action.***

## 3.9 Comparing the Futures

As stated, the future of the plutonium inventory is defined by:

1. A period of storage, during which the hazard represented by the $PuO_2$ powder reduces as improvements are made to the storage conditions. Even when this activity is complete the hazard remains high.
2. A period of manufacture, during which the material is progressively converted into a solid, lower hazard form, which is for either:
   a. *Disposal* in the UK's GDF, at a date still to be determined but currently envisaged to be in the decades beyond 2060, or
   b. *Use* as fuel, in reactors of a type not yet determined, but either as a one-off use in LWRs generating somewhere over 90 GWy of electricity, or as a precursor to a potentially much larger low-carbon energy yield in the nucleation phase of a yet-to-be-defined fast reactor programme.

The major programmes are illustrated in Figure 1. All these options involve lengthy periods of activity: the initial decades of reworking the inventory material and its storage; and a subsequent manufacturing campaign, also presumably of some decades, to turn the stored plutonium dioxide into either fuel for reactors, or into a wasteform for disposal in the UK's GDF. It follows that, provided only that it is accepted

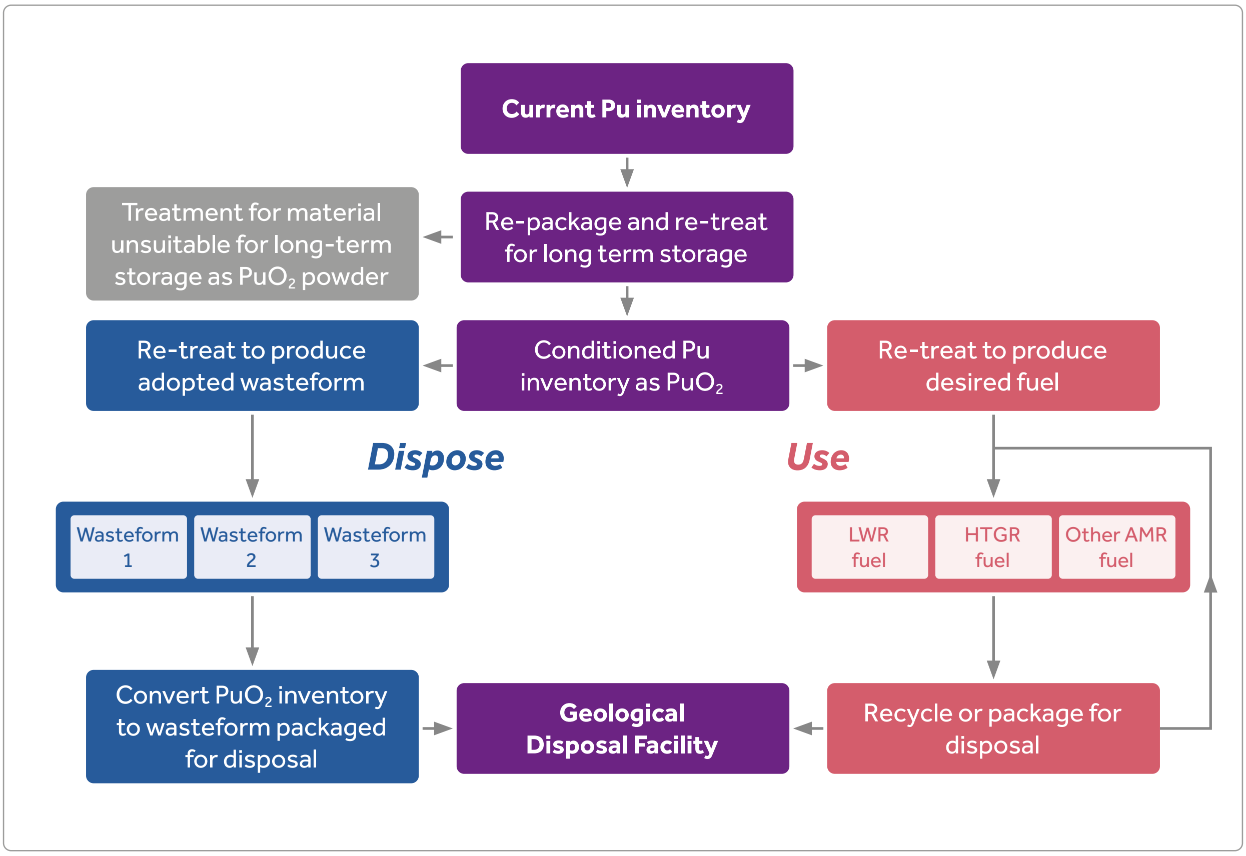

**Figure 1. Plutonium inventory flowsheet choices.**

that improving the condition and storage of the inventory is both essential and on the critical path, then there is no requirement for any short-term decision-making on the choice between *Use* and *Dispose*.

However, there is ample justification for a reasoned programme of work to examine the components of the two forward paths with the view to make a choice on the basis of an explicit assessment of the benefits, detriments and costs of the optimum variant of each path. This is perhaps, the central finding and recommendation of this study:

1. The urgent work, essential to the reduction of the hazard of $PuO_2$ storage has been identified, is underway, and must be prioritised and kept to programme.
2. The duration of this improvement activity (more than two decades) must be used to study the benefits and detriments of the *Use* and *Dispose* options, in the context of the ongoing need to achieve Net Zero in the long and very long term.
3. An additional factor must be the evaluation of the fate of the plutonium (and uranium) contained in the currently stored spent AGR and PWR fuel, which must be clarified before any action is taken to move to their disposal in the UK's GDF.

It is instructive to examine the effect of the forward paths on the assessed hazard of the UK plutonium system. The effect of these campaigns on the hazard associated with the inventory, as measured by SED (see Section 3.7 and Appendix 3) is illustrated in Figure 2. Note that the horizontal axis indicates the degree of processing in the campaign indicated, with some nine orders of magnitude decrease as the storage form and regime are optimised, and a further six or so orders of magnitude reduction as the manufacture of the solid waste or fuel form proceeds*.

This emphasises the importance of the inventory improvement activity and reinforces the major recommendation to set up significant programmes now

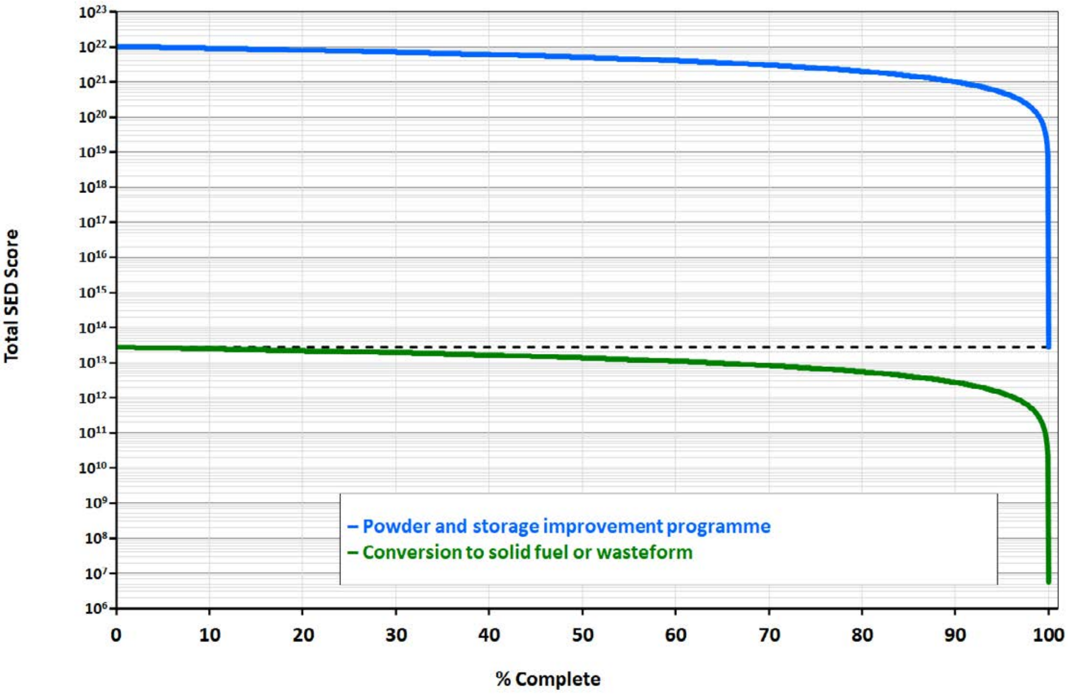

**Figure 2. Hazard evolution during inventory activities using SED measure.**

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* *Note that, because the SED scale is exponential, the effect of the improvement programme appears limited in its initial stages i.e. a reduction of 90% in the amount of* $PuO_2$ *stored in sub-optimum conditions will only decrease the SED score by one order of magnitude.*

to study the future options. The original intention of this study was to delve into the possible *Use* and *Dispose* programmes to derive critical path activities and estimated costs. However, at this stage the level of knowledge of these estimates would be limited by:

1. The centrality of the inventory improvement activity, and the relatively long timescale on which decisions are required.
2. The relatively undefined nature of the nuclear contribution to the UK's Net Zero campaign, and the size and type of future reactor programmes – this will give the background to the "plutonium carbon saving benefit" and its long- and short-term importance.
3. The unknowns in the *Dispose* programmes including the wasteform and the costs of wasteform production and disposal.

With this background, it is felt more relevant to include such estimates in the early stages of the study of future options.

The long times involved may be compared with the much shorter timescales over which nuclear policies and programmes have varied, changed or reversed in the past. However, the long interval before real change would occur is surely a major incentive to institute and maintain a holistic assessment of the possible futures, in particular to study the dependencies of any given future on earlier actions.

Thus the only policy agreement that is presently required is that the current programme of improvement to plutonium storage conditions is sensible, justified and will continue to be funded. This programme will then set the background timescale for the next steps in the inventory decision making, which should remove the need for short term decisions – apart from the instigation and funding of the holistic assessment programme discussed above.

***Recommendation nine:***

***The decades-long, highly challenging programme needed to address the challenges of the UK plutonium stockpile can only be delivered by an experienced community of practice so Government should ensure there is a sufficient supply of suitably qualified and experienced personnel to deliver the programme.***

***Recommendation ten:***

***Government should ensure that a robust, long-term programme of Research, Development and Innovation (RD&I) is in place to support selection and implementation of any plutonium management option.***

# 4

# Conclusions

There is ample evidence that Government has not – in recent decades – rushed into the fray of policy and decision making regarding the future of the UK plutonium inventory. There are several reasons for this:

- Resources within Government have been very stretched over recent years and continue to be stretched.
- Decisions around the future use and management of plutonium would ideally be made against a backdrop of clear understanding about future UK plans for nuclear new build (technology, timeframe, scale, fuel strategy, and so on). Such understanding has not yet been clarified, and so there remains a risk that decision made now about the future of plutonium stocks could be regretted in the future once these plans take shape.
- Timescales for the various options for managing the plutonium inventory are all many decades in duration, which creates a persuasive perception that important (and sometimes irrevocable) decisions are best delayed for a while until more information is available.
- Other nuclear and energy agendas in the context of net zero are more eye-catching and easier to present in a mainly positive and media-friendly light.
- Announcements about plutonium policy are likely to attract robust criticism and be misrepresented as either perpetuating risk or pre-judging policy decisions around future new build programmes.

Acknowledgement that the future management of plutonium is an unavoidably complex, multi-generational undertaking, requiring ongoing stewardship prior to an irrevocable decision on the end point for the material would be an important first step in moving the situation forward. Equally, recognition of the significant major uncertainties inherent in any future strategy is vital, and so there is clear value in a long term, programmatic approach with continuity, flexibility and adaptability.

Research, Development and Innovation (RD&I) can play a vital role to help decrease uncertainties and provide opportunities to accelerate the programme and reduce cost, in addition to developing the specialist skills needed to deliver a future programme – whatever that programme may look like.

### *Overarching Recommendation*

***Government, which is ultimately responsible for management of the UK's plutonium stockpile, should acknowledge that this is an unavoidably complex, multi-generational undertaking, requiring ongoing stewardship prior to an irrevocable decision on the end point for the material, and should put in place suitable arrangements. There are significant major uncertainties which can only be managed through a long term, programmatic approach with continuity, flexibility, adaptability, underpinned by Research, Development and Innovation (RD&I) commensurate with the scale of the challenge. This RD&I will not only decrease uncertainties and provide opportunities to accelerate the programme and reduce cost but will also be essential in developing the specialist community of practice required for delivery. The recently concluded Advanced Fuel Cycle Programme may be a useful model.***

# Recommendations

### Recommendation one

*Before attempting to make and implement policy decisions regarding plutonium, Government should ensure that a national dialogue takes place allowing stakeholders from all sides to share their views and participate in a respectful, evidence-based debate. This must be more than either a "lip service" consultation with outcomes already decided or a polarised "black versus white" argument. It needs to be an open dialogue, facilitated and led by trusted voices and based on a clear view of Government's thinking of the role (if any) plutonium might play in meeting future UK energy needs.*

### Recommendation two

*The current programme of repackaging and storing the plutonium inventory in optimal conditions must be carried out by the NDA and Sellafield Ltd to the currently programmed end point of 100-year design life storage. This provides sufficient time for the necessary comprehensive Research, Development and Innovation (RD&I) required to underpin the whole plutonium lifecycle to be carried out in parallel.*

### Recommendation three

*Bringing the UK plutonium programme to a successful end will take several decades at least. Both Government and NDA should provide the commitment and resources needed to ensure continuity and development of capability over this timescale.*

### Recommendation four

*Government should decide on and implement a preferred end point for the plutonium once a satisfactory assessment of options and their attributes is available, taking into account changes in storage environment and the hazard that plutonium presents.*

### Recommendation five

*Given the hazard represented by the plutonium stockpile and the long duration of plutonium storage, the storage infrastructure is critical to safety and security, so Government, NDA and other stakeholders must ensure that sufficient attention and resources are devoted to long term care of these assets.*

### Recommendation six

*The hazard represented by the plutonium stockpile would be greatly decreased by conversion from dispersible powder into a solid form, but the choice of form will determine which future option is to be followed. Government should ensure that a comprehensive assessment is carried out on the attributes and costs of the range of options.*

### Recommendation seven

*The different disposition options follow very different pathways to putting plutonium beyond reach. To underpin decision-making, Government needs to develop a full understanding of the whole plutonium lifecycle for each pathway before committing to irrevocable decisions.*

### Recommendation eight

*Because of the major uncertainties associated with the UK's plutonium management programme, it is unwise to rely on discounted costs to evaluate the programme and, in particular the assumption of cost decrease associated with discounting should not be used as a pretext to delay decision making and action.*

### Recommendation nine

*The decades-long, highly challenging programme needed to address the challenges of the UK plutonium stockpile can only be delivered by an experienced community of practice so Government should ensure there is a sufficient supply of suitably qualified and experienced personnel to deliver the programme.*

### Recommendation ten

*Government should ensure that a robust, long-term programme of Research, Development and Innovation (RD&I) is in place to support selection and implementation of any plutonium management option.*

### Overarching Recommendation

*Government, which is ultimately responsible for management of the UK's plutonium stockpile, should acknowledge that this is an unavoidably complex, multi-generational undertaking, requiring ongoing stewardship prior to an irrevocable decision on the end point for the material, and should put in place suitable arrangements. There are significant major uncertainties which can only be managed through a long term, programmatic approach with continuity, flexibility, adaptability, underpinned by Research, Development and Innovation (RD&I) commensurate with the scale of the challenge. This RD&I will not only decrease uncertainties and provide opportunities to accelerate the programme and reduce cost but will also be essential in developing the specialist community of practice required for delivery. The recently concluded Advanced Fuel Cycle Programme may be a useful model.*

# Appendix 1: The UK's Plutonium Stockpile

## Origin of the Material

The UK has manufactured plutonium, initially for military purposes, since 1950. Subsequent reprocessing of civil fuel was intended to provide a reserve of plutonium to support the UK fast reactor programme, but separation continued even after that ended in 1994. The continuing use of uranium metal fuel in the first-generation Magnox reactors obliged the UK to carry on reprocessing, whereas the decision to reprocess oxide fuel in THORP was a commercially-driven choice. As a result, the UK presently holds the world's largest stockpile of separated civil plutonium (around 140 t). While the history of the reprocessing programme is complex and much information is not in the public domain, some is available. There are three distinct subsets of material though clearly, within these, there are further smaller differences:

- Plutonium from UK power reactors,
- Plutonium from overseas fuel, and
- Plutonium from the fast reactor programme.

An overall timeline of the stockpile amounts from varying sources is shown in Figure 3 along with key milestones in the UK's reprocessing operations.

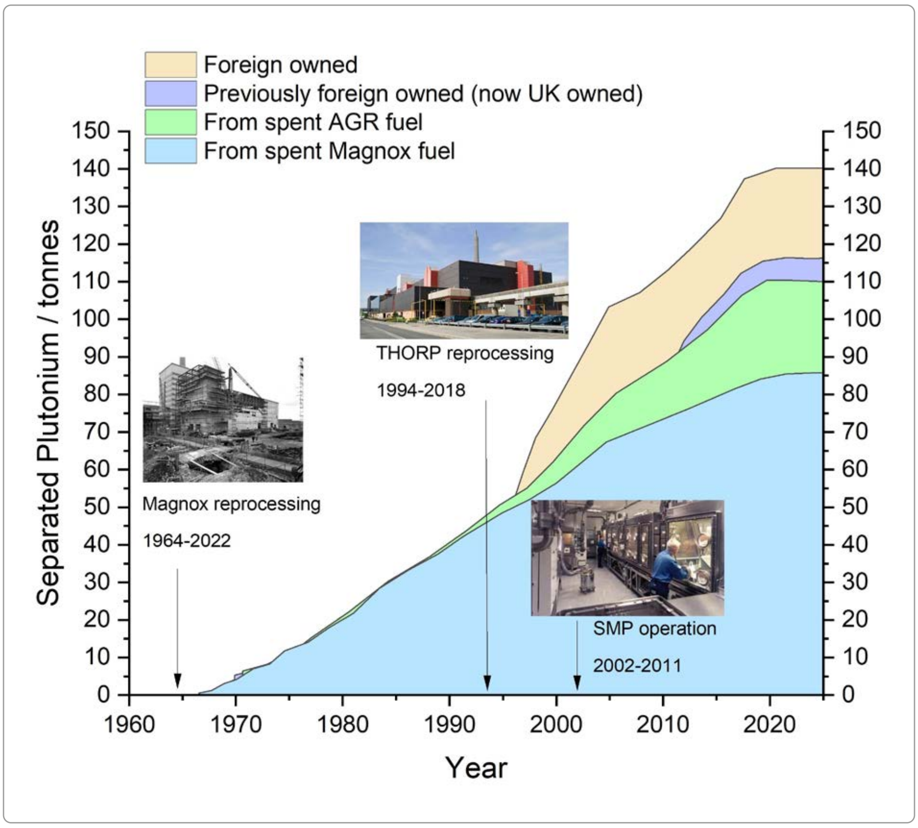

**Figure 3. Timeline of UK stockpile of civilian separated plutonium from different origins reproduced from [14, Fig. 1], along with key milestones in the UK's reprocessing operations.**

## Plutonium from UK Power Reactors

Plutonium from domestic power reactors constitutes the majority of the UK's stockpile. 85.8 t originate from Magnox reprocessing between 1964 and 2022 and is therefore derived from low burnup (3-5 GWd $t^{-1}$) natural uranium fuel. Due to the low fuel burnup of Magnox reactors, this material consists mainly of $^{239}$Pu and $^{240}$Pu, with a lower $^{241}$Pu isotopic content, making it more attractive for use as MOX fuel feedstock due to lower gamma radiation fields from ingrown $^{241}$Am. However, due to the use of PolyVinyl Chloride (PVC) in some storage containers and its subsequent degradation, ~5 t of this material is contaminated with chlorine which makes it more difficult to manufacture into nuclear fuel since the chlorine will need to be removed before manufacture.

23.6 t originates from THORP reprocessing of Low Enriched Uranium (LEU) AGR fuel (medium burnup; 18-40 GWd $t^{-1}$) between 1994 and 2018. Due to the higher fuel burnup in AGRs compared to Magnox, this material contains higher amounts of $^{241}$Pu which, when stored for long periods, results in increased amounts of $^{241}$Am ingrowth, which renders the plutonium less attractive for MOX fuel manufacture.

A series of relatively small transfers of foreign-owned plutonium reprocessed at Sellafield was carried out between 2012 and 2014 to simplify arrangements. The UK took ownership of 4.75 t of German, 1.85 t of French, 0.8 t of Swedish, 0.6 t of Spanish and 0.35 t of Dutch plutonium. Some of this overseas material will be derived from Magnox reprocessed fuel (from Tokai Unit 1 and Latina), but most derives from LEU oxide fuel irradiated in LWRs (40-60 GWd $t^{-1}$), so the majority makes up additional THORP reprocessed material.

In total, the UK owns ~116.5 t [15, p. 3] of separated civil $PuO_2$ although accurate inventories are complicated by historical transfers between civil and military stockpiles, and these may account for discrepancies between different estimates [16].

Although outside of the scope of this paper, the Office for Nuclear Regulation (ONR) also estimates that a further ~27 t of plutonium contained within irradiated spent fuel is held mainly at nuclear power stations and reprocessing plants and this quantity will continue to increase with time [15, p. 4].

## Plutonium from Overseas Fuel

Significant quantities of overseas fuel were reprocessed at Sellafield with the intent to manufacture into MOX fuel and return to overseas customers. With the closure of the UK's MOX manufacturing plant in 2011, most of the plutonium separated is still stored on Sellafield site but the overseas utility companies remain the owners and are ultimately responsible for this material. Currently, there are 24.1 t which are owned by other countries [15, p. 3], mostly Japan (21.8 t), produced from THORP reprocessing. A transfer of 0.65 t stored at Sellafield was made from German to Japanese ownership. These materials are managed in line with any contractual commitments from the foreign customer and are stored by the NDA on the Sellafield site.

## Plutonium from the Fast Reactor Programme

Fast reactors use driver fuel which has a high fissile content and provides the surplus neutrons which convert fertile $^{238}$U in the breeder elements into fissile material. The Dounreay Prototype Fast Reactor (PFR) used MOX driver fuel with a plutonium content of 20-33% [17, 18, Fig. 5.3.3] while irradiated breeder material will also contain plutonium. In total around 100 t of fuel were still at Dounreay in 2013, of which about 26 t was "exotic" (i.e. high fissile content material containing plutonium or highly enriched uranium). In total, 13 t of this exotic material contained plutonium [19, p. 11], with a total inventory of around 2 t, comprising metal, carbide and oxide. 44 t of the total were irradiated breeder material.

The irradiated breeder elements have been transported to Sellafield and reprocessed through the Magnox reprocessing plant, so plutonium from that source is now included in the Magnox product described above. In terms of the total UK stockpile, the exotic material is small but some components, notably metal and carbide, are chemically reactive and may be challenging to manage.

## Comparison with Other Nations

It is difficult to find data which allow direct comparison of national holdings of fissile materials. Table 2 (page 23) summarises the main holdings declared to the International Atomic Energy Agency (IAEA) as of 31st Dec 2021.

The UK stockpile has a number of unique characteristics in comparison with the other large holdings. Military material is generally derived from low burnup fuel and therefore has a high $^{239}$Pu content, typically >93 at%, with most of the balance being $^{240}$Pu. Heat output (~2.2 W $kg^{-1}$) and $^{241}$Am ingrowth are significantly lower than the UK stockpile material. Moreover, much military material will be stored as metal, not oxide. The French civil stockpile, while large and similar in isotopic composition and form to the UK's, is younger since the Melox MOX plant uses plutonium within five years of separation to limit $^{241}$Am content, and this will also limit the effects of any other ageing phenomena.

**Table 2. Principal global plutonium holdings by country (tonnes).**

| Country | Military | Civil |
|---|---|---|
| Russia [20] | 88.0 | 103.1 of which the main elements are 57.0 at reprocessing facilities and 40 of excess weapons material. |
| USA [21] | 38.4 | 49.4 of which the majority (41.3) is excess weapons plutonium originally intended for use in MOX fuel, though this seems less certain now. |
| France [22] | 6.0 | 84.9 largely for recycle in MOX fuel. France holds an additional 15.0 tonnes of separated plutonium plutonium belonging to other nations. |
| China* [23] | 2.9 | 0.04 |
| UK [15] | 3.2 | 116.5 plus an additional 24.1 tonnes of separated plutonium belonging to other nations. |
| India [24] | 0.7 | 8.5 |
| Japan [25] | 0.0 | 45.8 comprising 9.3 in Japan, 21.8 stored in the UK and 14.7 stored in France. |

** As at 31st Dec 2016.*

## Storage of UK Plutonium

Plutonium materials are stored in robust metal cans which each contain a maximum of a few kilograms of material; in total, there are over 30,000 cans [26, p. 11]. Modern storage is based on a sealed can-in-can arrangement to provide multiple layers of containment. Earlier arrangements included some breathable cans which allowed outgassing through a filter, and some packages which included a layer of non-metallic material, notably PVC as mentioned above.

Earlier cans were not intended to store material for as long as has been the case. The assumption was that the plutonium would be reused in fast reactor fuel within a couple of decades which has not happened. It has become clear that, over decades in storage, $PuO_2$ reacts with moisture and with components of the can [27]. These processes are not yet fully understood but lead to changes in the pressure within the can and to alteration of the plutonium material itself. Since there is a relatively large mass of $PuO_2$ in the can, the majority will remain unaltered but there is obvious potential, for example, for these processes to modify the $PuO_2$ surface.

Additionally, in some of the early cans that used PVC, the PVC has degraded, leading to corrosion of the cans and contamination of the plutonium with chlorine [28]. While considerable work has been undertaken to stabilise this material, around 5.5 t in total is not immediately suitable for fuel manufacture, without prior removal of the chlorine. Some cans currently measure >100× the allowable chlorine content for the MOX fuel specification [29] and would need treatment to reduce these levels, which could be achieved through thermal treatments [28]. A further 6 t of material is deemed out of specification for MOX fuel fabrication due to low specific surface area which again could be improved by thermal treatments similar to those used in uranium oxide fuel recycling. Further understanding is clearly needed of these out-of-specification batches to understand effects of further ongoing storage of these materials. Additionally, the development of re-treatment and packaging procedures and eventually inform fuel and/or wasteform manufacture processes, plant design and product specification (which is much less well-defined for the disposal wasteform).

A major new facility, the Sellafield Product and Residues Store Retreatment Plant (SRP), will be used to retrieve, re-treat (e.g. remove high levels of chlorine contamination) and repackage the plutonium into welded stainless steel canisters designed as 100-year packages [30]. These will then be transferred to a new storage facility within the Sellafield Product and Residue Store (SPRS) to ensure safe and secure storage of the stockpile into the next century until a pathway to a permanent end point can be identified. Current estimates expect plant commissioning in the 2020s [30, p. 6] and repackaging operations to be completed by ~2060 [31, p. 140].

## UK Experience of MOX Manufacture

The United Kingdom Atomic Energy Authority (UKAEA) production division, which later became British Nuclear Fuels Ltd (BNFL), began fabrication and testing of MOX fuels in the 1960s. This produced ~17 t of fast reactor fuel for the Prototype Fast Reactor (PFR) at the Dounreay site which went critical in 1974. Additionally, a further 3 t of thermal reactor MOX for irradiations in Europe were produced [32]. Subsequently, BNFL began developing the SBR process for MOX fuel manufacture in the 1980s [33] that was successfully demonstrated in irradiation experiments in the US in water cooled reactors. Following this, the MOX Demonstration Facility (MDF) began operation in 1993 as a pilot plant to provide customers such as Germany, Japan and Sweden with fuel elements that would conform to the customers' and regulators' requirements, with a larger commercial plant, the Sellafield MOX Plant (SMP), planned for scale up [34]. Although it produced more than 10 t of MOX fuel in total [35], operations at the MDF were suspended in 1999 due to the discovery of data falsification from a second manual quality check of MOX pellets that were en-route to Japan and also resulted in the removal of MOX fuel rods fabricated at MDF from German reactors where data duplication was also found [32].

The UK then operated SMP between 2002 and 2011 with the intention to fabricate MOX from the UK and foreign-owned plutonium stockpiles for use in LWRs. The plant was designed to manufacture fuel from THORP grade plutonium, with a projected annual throughput of 120 tHM. However, by 2008 the plant's actual production capability was quoted as 5-7 tHM per year or 15 tHM with modifications. Ultimately, with the throughput difficulties (partly due to a failed BNFL acquisition of Siemens MOX expertise leading to gaps in design and operational capability), and the high number of Japanese utility contracts affected by the Fukushima accident, the NDA concluded that SMP should close to limit the financial risk to the UK taxpayer. During its operating lifetime the SMP manufactured around 13.8 tHM of MOX fuel [36].

## Behaviour of Plutonium

Given the different origins of the material, the isotopic composition of separated plutonium from different sources will vary (Table 3).

Furthermore, radioactive decay will change the composition and nature of plutonium materials progressively over time (Figure 4). Alpha decay processes produce isotopes of uranium, all of which have relatively long half-lives (>245,000 years). From a proliferation perspective, $^{239}$Pu decays to $^{235}$U, also a fissile isotope, which has a half-life of 704 million years, so decay alone will not eliminate the possibility of proliferation.

The β-decay of $^{241}$Pu leads to relatively rapid, in the context of decades-long storage of plutonium, production of $^{241}$Am, which decays by α-emission with a 433-year half-life, in turn forming $^{237}$Np which, since it has a half-life of 2.14 million years, effectively terminates this decay chain. The ingrowth of americium has practical significance since its α-decay is accompanied by emission of a 59.5 keV γ-photon at 36% yield, leading to significant gamma doses from aged plutonium materials.

**Table 3. Isotopic composition (at%) and decay heat output (W kg$^{-1}$) at the time of separation for plutonium of different origins (based on [37]).**

| | $^{238}$Pu | $^{239}$Pu | $^{240}$Pu | $^{241}$Pu | $^{242}$Pu | Decay Heat |
|---|---|---|---|---|---|---|
| Half-life (yr) | 87.7 | $2.4\times10^4$ | $6.5\times10^3$ | 14.4 | $3.8\times10^5$ | |
| Decay type | α | α | α | β | α | |
| Magnox 3 GWd t$^{-1}$ | 0.10 | 80.00 | 16.90 | 2.70 | 0.30 | 3.3 |
| Magnox 5 GWd t$^{-1}$ | n/a | 68.50 | 25.00 | 5.30 | 1.20 | 3.2 |
| AGR 18 GWd t$^{-1}$ | 0.60 | 53.70 | 30.80 | 9.90 | 5.00 | 6.9 |
| LWR 33 GWd t$^{-1}$ | 1.30 | 56.60 | 23.20 | 13.90 | 4.70 | 11.0 |
| LWR 53 GWd t$^{-1}$ | 2.70 | 50.40 | 24.10 | 15.20 | 7.10 | 17.7 |

In addition to the radioactive decay products, α-decay also produces $^4$He. Some of the helium is retained within the $PuO_2$ crystal structure and some is released into the container. Helium production rates from $PuO_2$ are in the range 0.004 to 0.014 ml $y^{-1}$ $g^{-1}$. The decay process will also damage the crystal structure of the $PuO_2$ (see Appendix 4) while decay heating can lead to centreline temperatures in the cans of up to 400°C.

## Risks

Plutonium is sometimes said to be "the most toxic substance known to man" (e.g. [38]). This partly reflects mystique associated with its origins and use in nuclear weapons programmes, and partly the genuine hazards it represents. Without debating definitions of toxicity at length, it is undoubtedly true that plutonium is a very hazardous material with the potential to cause harm through a variety of processes. The material from the Magnox and THORP reprocessing operations is stored as $PuO_2$ powder in the micron particle size range, the majority below 45 μm diameter [28]. This represents a significant fraction of "fines" in the stockpile (i.e. particles which can be aerosolised and spread easily in air in the event of a release of material or lead to contamination of plant parts such as gloveboxes when handled).

## Security and Safeguards

Because of the health, environmental and proliferation risks plutonium presents, it is kept in very secure conditions. It is stored behind multiple layers of physical protection, with access heavily restricted. The costs of securing the UK plutonium stockpile are currently around £73 million per year, while periodic upgrading or replacement of plutonium stores costs £ hundreds of millions [39].

Except for military material, all UK plutonium is subject to international safeguards [40]. These are ultimately overseen by the IAEA, an arm of the United Nations, and allow the UK to demonstrate that its nuclear activities are consistent with the commitments it has given under international agreements.

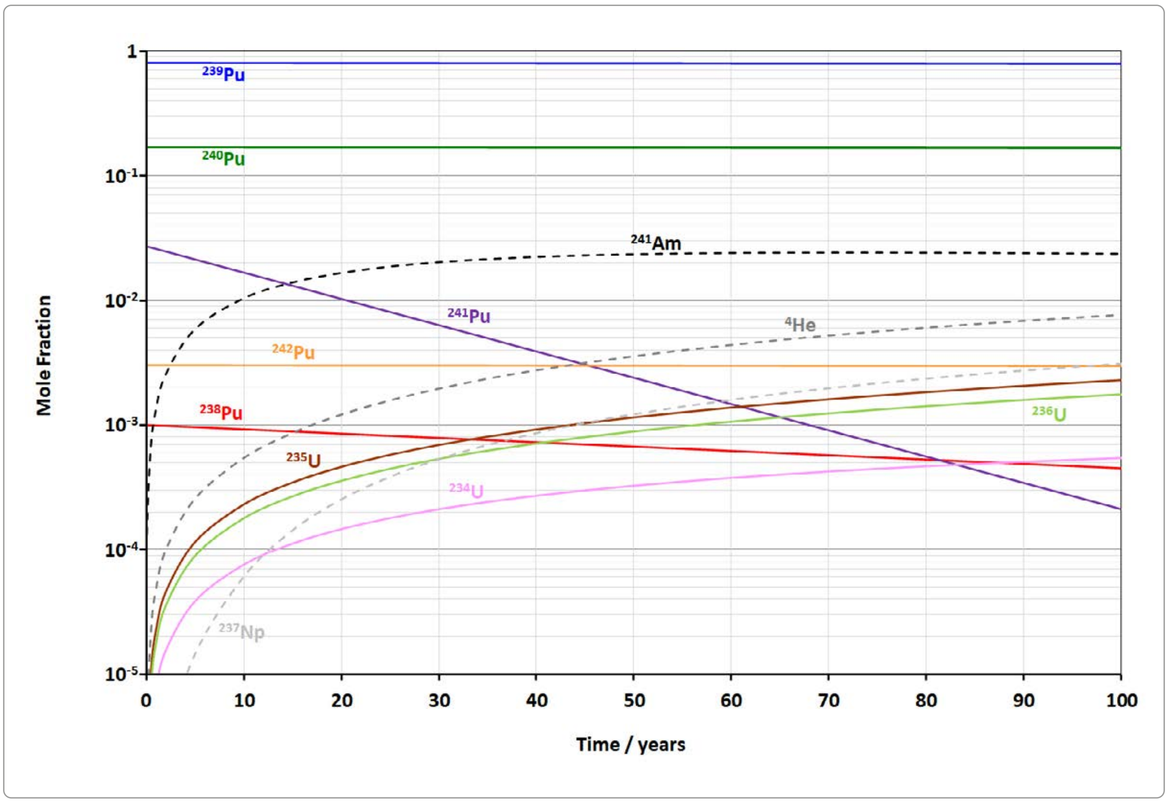

**Figure 4. Compositional change in Magnox (3 GWd/tHM) plutonium-bearing materials due to radioactive decay as a function of time.**

## Health and Environment

Plutonium materials do not represent a major external radiation hazard since the common plutonium isotopes do not have strong γ-emissions. Older plutonium, with significant $^{241}$Am present, can represent a significant external radiation hazard, especially to workers handling large quantities because of its strong gamma emission.

Neutron radiation may be a problem in some circumstances. Some plutonium isotopes have a reasonably high probability of spontaneous fission decay, with $^{240}$Pu being the most important in the stockpile. Spontaneous fission events number 471 $s^{-1}$ $g^{-1}$ in $^{240}$Pu and, in plutonium materials containing other elements, the neutron background can be further increased by (α,n) reactions, a particularly important phenomenon in compounds such as $PuF_3$ or $PuF_4$.

The principal risk from plutonium materials arises from internal contamination through contaminated wounds or, particularly, inhalation (it is inefficiently transferred across the gut so ingestion is less problematic, though still ill-advised). Inhaled plutonium translocates from the lung mainly to bones, bone marrow and liver, and is only slowly excreted from the body.

The specific impact from an inhalation exposure will depend on many factors including the airborne release fraction, particle size, chemical form and deposition efficiency in the lung, which determine the absorption rate into the blood or clearance rate into the digestive tract. Inhalation of plutonium in quantities of the order of a few hundred becquerels, corresponding to a mass in the sub-microgram range, will lead to significant exposures or overexposures. Plutonium materials, particularly powders like $PuO_2$ such as those that make up the majority of the UK's stockpile, are therefore handled in highly engineered facilities with strict safety procedures and precautions against accidents that could cause exposure particularly via inhalation. Stringent precautions are taken to avoid accidental releases and plutonium stocks are held under very tight security to prevent malicious use and under these conditions, exposure to workers and the general public has been and will be insignificant [41, Sec. 3.1].

## Criticality

A criticality accident is the unintended initiation of a nuclear fission reaction. Such an event will not lead to a nuclear weapon-like explosion because the material will disassemble much too rapidly but will lead to very dangerous radiation and neutron doses and can be difficult to control. The factors influencing the likelihood of a criticality accident include the quantity of fissile material present, its geometry (since this controls neutron leakage), and the presence of moderating, reflecting and neutron-absorbing materials.

Stockpile plutonium materials present a significant criticality risk [42]. Criticality safety can, however, be assured by carefully limiting quantities of material, using safe geometry vessels such as annular or "pencil" tanks, and including criticality safety in materials selection, possibly even deliberately adding neutron-absorbing poisons.

In geological disposal, plutonium may present some unique challenges with respect to criticality [43]. In a uranium-driven criticality the associated temperature rise decreases the reactivity of the fissile material and, in the absence of a further supply of fissile material, for example a continuing influx in groundwater, the reaction will cease. If there is a continued supply of fissile material, then the reaction may continue over a long period of time (i.e. millennia) with only limited temperature increase (albeit up to a few hundred degrees Celsius) and only very localised effects. This situation is a "quasi-steady state" criticality and is essentially the one which pertained in the Oklo natural fission reactors [44].

Although a plutonium-driven criticality is also likely to be a quasi-steady state event, in some highly unlikely circumstances, the temperature increase from a plutonium-driven criticality may actually increase the reactivity of the fissile material, leading to acceleration of the reaction and a rapid rise in temperature and pressure, a "fast transient" criticality. The duration of such an event would be less than one second, but it would cause physical damage, for example fracturing the surrounding rock. It should be appreciated that a fast transient event requires a very specific set of circumstances and is therefore felt not to be credible in GDF conditions.

# Appendix 2: Options for Plutonium Management

## The Plutonium Dilemma

The UK has substantial inventories of fissionable, fissile and fertile materials[*] for civilian purposes. The principal components are separated plutonium and Depleted, Natural and Low-Enriched Uranium (DNLEU). The plutonium stockpile is discussed in detail in Appendix 1 while, in the 2019 Radioactive Materials Inventory, the UK stockpile of DNLEU is given as 99,000 tonnes [45]. If completely fissioned, the plutonium reserve corresponds to approximately 3,000 TWh[†] and the DNLEU to around 2,200,000 TWh. To put these in context, UK final energy consumption is around 1,600 TWh per year [46, p. 10], so these materials could potentially meet the UK's energy needs for centuries. However, the many technical, safety, security and societal obstacles to extracting this energy from the materials stockpiles may be insurmountable. This leads to a range of options for managing the UK's nuclear materials, which we summarise here.

## Disposal as Waste

Ostensibly, the quickest and cleanest way of dealing with the plutonium stockpile would be to manage it as waste. The UK is making progress towards a GDF to accommodate the Higher Activity Waste[‡] inventory. The GDF will be built 200 to 1,000 m below the surface in a suitable geological setting. Plutonium would be disposed as part of the ILW component, which is estimated at 499,000 $m^3$ packaged volume. Plutonium would only be a small addition to this volume, probably less than 5,000 $m^3$. It is obvious that disposal of plutonium as waste would sacrifice its energy content and almost certainly preclude the UK from adopting fast reactors (see below). That said, as described earlier, about 6 t of the plutonium stockpile have become chlorine-contaminated in storage due to deterioration of PVC packaging, and the end point options for such material are not clear. Whether it is used as fuel or fabricated into a wasteform, the product specification must either allow for a relatively high chlorine content, or the material will have to be reworked to reduce the chlorine content.

While the preferred wasteform for plutonium disposition has not yet been identified, several options are available, including disposal MOX (a mixed uranium/plutonium oxide optimised for disposal), single phase[§] and mixed phase ceramics (e.g. Synroc), and composite materials such as glass-ceramics [47]. For disposal of plutonium, the wasteforms may well incorporate a neutron-absorbing poison such as gadolinium to limit the possibility of post-disposal criticality. A clear specification for any plutonium wasteform would be needed, together with development work to underpin the required industrial scale manufacturing process.

Whichever wasteform is finally chosen, the implementer of geological disposal would have to carry out a full disposability assessment to provide confidence that plutonium disposal will not breach the GDF environmental safety case. The potential for post-closure criticality, arising from physical/chemical redistribution and accumulation of plutonium-rich material, is a particular concern. Given the stakeholder concerns which plutonium inevitably attracts, geological disposal of large quantities of plutonium would require

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[*] *A **fissionable material** is a material where there is some unspecified neutron energy at which the probability of fission is non-zero; **fissile materials** are a subset of fissionable materials which undergo fission with thermal neutrons; **fertile material** can be converted into fissile material in a nuclear reactor through neutron capture and beta decay reactions. Thus, $^{235}U$ is a fissile (and hence also a fissionable) material, whereas $^{238}U$ is fissionable but not fissile (since it only undergoes fission with fast neutrons) but is also fertile since it can be converted into $^{239}Pu$. All actinide nuclides are fissionable with neutrons with energies >1MeV.*

[†] *Some references use the energy unit "million tonnes of oil equivalent" (Mtoe), where 1 Mtoe = 11.63 TWh. All instances in this document have been converted to TWh for clarity.*

[‡] ***Higher Activity Waste** comprises High Level Waste, Intermediate Level Waste and a small portion of Low Level Waste. Unreprocessed spent fuel will also be consigned to the GDF and, if managed as wastes, then nuclear materials will also be disposed to it.*

[§] *Many candidate phases have been proposed including zirconolite ($CaZrTi_2O_7$), sodalite ($Na_8(Al_6Si_6O_{24})Cl_2$), pyrochlore ($(Na,Ca)_2Nb_2O_6(OH,F)$) and perovskite ($CaTiO_3$).*

strong stakeholder engagement and clear communication. Transport, security, retrievability and the potential for a bad actor to obtain fissile material in the future are likely to be prominent concerns. In assessing the alternative of reuse of the separated plutonium, it is also necessary to compare the direct disposal of the separated plutonium and the disposal of spent fuel from reuse.

## Use in Thermal Fission Reactors

The use of plutonium in thermal reactors, as MOX fuel, is well established. However, the presence of resonances for $^{239}$Pu and $^{241}$Pu in the thermal neutron spectrum adds positive terms to the reactivity coefficient with temperature, requiring consideration in the safety case. Plutonium, often at 5-8 wt%, can be mixed with depleted uranium and fabricated into fuel for use in light water reactors, often at a core loading of about 33% although the regulatory assessment of the EPR in the UK contemplates up to 50% and the EPR can operate with a 100% MOX core [48]. However, EDF Energy have stated clearly that "Mixed Oxide (MOX) fuel is not being considered as a fuel source for Hinkley Point C (HPC) at this point in time" [9, p. 1] and it is not clear whether or not the Hinkley Point C reactors are actually being configured to allow MOX use or whether they would require modification. Furthermore, the regulatory justification for Hinkley Point C explicitly does not consider the use of MOX fuel [49]. The presumption must therefore be that MOX is not a serious consideration for the UK EPR fleet. The NDA also considered using a modified CANDU heavy water moderated reactor design to irradiate MOX fuel and convert separated plutonium to spent fuel. This option was still considered credible in 2014 [50].

As with uranium fuels, the spent MOX can either be disposed as waste or, in principle, can be recycled, although multi-recycled plutonium brings some challenges due to shifts in isotopic composition. Only the odd mass numbered plutonium isotopes are fissile so, as plutonium is recycled multiple times, the even isotopes become relatively more abundant (Table 4), to the point that they would be problematic in accident conditions. There is thus a practical limit to the recyclability of plutonium, albeit that multi-recycle will reduce the fissile content and hence the proliferation risk. Also, the impact on reducing uranium consumption is small for a single recycle of MOX because of low conversion in a PWR. Each MOX recycle would produce the equivalent of around 1/7 of a PWR fuel loading. These considerations are not so relevant to fast reactors, as all plutonium nuclides have significant fission cross-sections, greater than capture cross-sections, for neutron energies >500 keV.

Once-through use of MOX fuel is very similar to once-through use of UOx. Both fuel types will have a significant plutonium content, although the MOX plutonium isotopic composition will be shifted to a lower fissile content (from Table 4, above, 68.8% fissile in first generation plutonium, decreasing to 56.81% in second generation material). Both fuel types will require a very long period of cooling, both thermally and radioactively, in interim storage before final disposal in a GDF.

An alternative way of using thermal reactors is to use the plutonium to fuel an advanced thermal reactor. Two options are with:

- High temperature reactors using either helium coolant (in the case of HTGRs) or fluoride molten salt coolant (Fluoride High-temperature Reactor; FHR), or
- A molten salt reactor with the plutonium as part of the fuel salt.

High temperature reactors use TRISO particle fuels that can be taken to very high burnup. Work in the USA evaluated "deep burn", with very high fractions of plutonium [51, 52]. There are various configurations with or without other minor actinides added to the plutonium, e.g. just 0.2% uranium added to the plutonium; with around 30% uranium added to the plutonium; thorium added to the plutonium; and "seed and breed" where some particles have a high concentration of plutonium and some just thorium. Irradiations in high flux materials test reactors have shown such high fissile content TRISO particles can be taken to over 700 GWd/tHM. These fuels are not easy to reprocess and would presumably be incorporated into a wasteform with minimal treatment to

**Table 4. Isotopic abundance of first and second generation plutionium isotopes from 33 GWd/t Uranium Oxide (UOx).**

| | $^{238}$Pu (at%) | $^{239}$Pu (at%) | $^{240}$Pu (at%) | $^{241}$Pu (at%) | $^{242}$Pu (at%) |
|---|---|---|---|---|---|
| 1st generation plutonium from 33 GWd/t UOx | 1.85 | 58.05 | 22.05 | 10.75 | 5.60 |
| 2nd generation plutonium from 33 GWd/t UOx | 2.74 | 42.51 | 29.19 | 14.30 | 9.82 |

remove excess graphite but keep the high integrity particles intact. This option would probably be the most effective in fulfilling safeguards requirements.

"Fuel-in-salt" molten salt reactors are less developed with only the Oak Ridge National Laboratory's Molten Salt Reactor Experiment (MSRE) providing operational experience. There is a range of configurations so it is worth just noting the option here without exploring all the possibilities [53, 54]. It should also be noted that the possibility of on-line chemistry (removing fission products and adjusting fissile content) offers the potential for both complete burning of plutonium and safeguarding problems, particularly if plutonium burning is combined with thorium in the salt, as removal of $^{233}$Pa (the precursor of $^{233}$U), could result in a very pure material usable in weapons [55].

## Use in Fast Fission Reactors

The big attraction of fast reactors is that they allow access to the huge energy resources presented by the plutonium, and particularly the DNLEU, stockpiles. Without fast reactors, the only use for $^{238}$U is slow, inefficient conversion into $^{239}$Pu in thermal reactors.

The plutonium stockpile, as a substantial reserve of fissile material, would be valuable if fast reactor technology were to be adopted. Because the reactor relies on fast neutron fission and fast neutron fission of $^{235}$U is comparatively inefficient, a plutonium driver fuel is attractive. The alternative is high assay uranium fuel (approaching 20% $^{235}$U enrichment). Even in these circumstances, doubling times would be of the order of 10-30 years, particularly if a radial blanket is avoided because of safeguarding considerations. A fast reactor would require 5-7.5 t/GWe for the first fuel load [56] and about three fuel loads would be needed for a single reactor to settle into the closed cycle. Overall, the amount per reactor is then 15 to 22.5 t/GWe, which implies the UK plutonium stockpile could support development of 6-10 GWe of fast reactor capacity, with a doubling time of ten years or more. Reactor design and the fuel material selected can reduce doubling time but at the cost of, for example, using more problematic fuel materials such as metal alloys. Breeding large quantities of plutonium in a "plutonium economy" raises significant security and proliferation concerns and there is increasing interest in using fast reactors as "burners", where they destroy more fissile material than they create, or "iso-breeders", where the quantity of fissile material is in steady state.

A fast reactor programme requires industrial scale fuel reprocessing, the fabrication and multiple recycling of tonne quantities of plutonium materials and brings with it major – but not insurmountable – safety and security challenges. While the UK has withdrawn from commercial reprocessing, it would be possible, with significant commitment and investment, to re-establish a closed fuel cycle capability and possibly a fast reactor programme. However, such a decision would need to be justified on grounds of energy security [57] and/or economics (compared to an open cycle, the closed cycle requires a substantial increase in uranium price and also a much longer time to realise the full economic benefits) [58]. In such circumstances, the current plutonium stockpile would be an asset, and the current enactment of a 100-year design life storage would keep the fast reactor option open for a credible timescale.

An alternative to the closed fuel cycle is to use fast reactors without reprocessing, as a means of returning the plutonium to the spent fuel state with some benefit from electricity generation. To avoid generating more plutonium the fast reactor characteristics can be adjusted to a burning rather than a breeding mode. The US metal fuelled Sodium Fast Reactor (SFR) "PRISM" was proposed by GE-Hitachi as one of the solutions to plutonium disposition by the NDA [59]. It was argued that the use of the hard neutron spectrum in a metal fuelled reactor would have advantages over an oxide fuelled SFR. There are advantages, but they are minimal because of the larger amount of inelastic scattering of neutrons in such reactors. Also there are disadvantages as they have less negative Doppler coefficients of reactivity and higher void coefficients. After the last review (in 2014) of the NDA project to assess the possibility of using the MOX version of CANDU or PRISM, the promised second stage of the review due to be published in 2017, did not take place. Proposals have also been made to use MOX fuelled SFRs for plutonium management [60]; the use of a small MOX fuelled SFR would have the advantage of being able to use UK capability and experience from PFR.

An extreme example of using reactors to burn plutonium and minor actinides, would be with "inert matrix" fuel, eliminating fertile uranium and thorium. A wide range of components and configurations have been explored. Examples include oxides of low neutron absorption elements like aluminium, magnesium, zirconium and cerium. Cerium has the advantage of being reasonably similar to uranium and plutonium in mixed oxides. Metal alloys, nitrides, carbides and silicides have also been explored [61]. Both thermal and fast reactors can be used but fast flux reactors have significantly higher neutron efficiency as neutron capture is much lower. Inert matrix fuels could be part of a reactor fuel loading, or they could be used in dedicated reactors, which would require the use of burnable poisons such as $^{10}$B, gadolinium, dysprosium, holmium, erbium or europium. The use of the rare earth elements also has a beneficial effect in improving the Doppler coefficient of reactivity, which is made less negative in the absence of the fertile nuclides $^{232}$Th and $^{238}$U.

## Use as a Neutron Multiplier in Fusion Reactors

The neutron economy in a reactor is critical to successful practical implementation of fusion power. The key processes in the fusion fuel cycle are production of energy from the fusion of deuterium ($^2H$) and tritium ($^3H$), shown in Reaction 1, which produces one neutron (n). The tritium, being a relatively short-lived radioactive isotope (half-life 12.3 years), has to be made from the $^6Li$ nuclide of lithium, in a reaction which consumes one neutron (Reaction 2). Both reactions release useful energy.

$$ {}^2_1H + {}^3_1H \rightarrow {}^4_2He + {}^1_0n + 17.6\ MeV \qquad (1)$$

$$ {}^6_3Li + {}^1_0n \rightarrow {}^3_1H + {}^4_2He + 4.8\ MeV \qquad (2)$$

Thus, to achieve balance, every neutron produced in a fusion reaction must also be used to produce a tritium atom. This is implausible since there will inevitably be some neutron loss from a fusion reactor through absorption in structural components or escape from the reactor. Moreover, the majority of the energy produced by deuterium-tritium fusion is associated with the neutron (14.1 MeV out of a total energy yield of 17.6 MeV), so that energy needs to be harvested efficiently. In the absence of any supplementary neutrons, the neutron economy of a fusion reactor will be precarious.

It is therefore necessary to include neutron multiplication in the tritium breeding component of a fusion reactor. Elements such as beryllium or lead can react with neutrons at the energies encountered in fusion reactors to produce additional neutrons (Reactions 3 and 4). However, these are threshold reactions, so they only occur above a threshold neutron energy and are endoergic (absorb energy).

$$ {}^9_4Be + {}^1_0n \rightarrow 2{}^4_2He + 2{}^1_0n - 1.6\ MeV \qquad (3)$$

$$ {}^{208}_{82}Pb + {}^1_0n \rightarrow {}^{207}_{82}Pb + 2{}^1_0n - 7.4\ MeV \qquad (4)$$

Neutron yields for fast neutron fission of fissionable materials are much higher than for (n,2n) reactions and increase significantly for neutron energies above 10 MeV. For example $^{239}Pu$ produces ~4.6 neutrons per fission from 14.1 MeV neutrons. In addition, the fission process itself is a substantial energy multiplier. Using the fusion neutrons in this way would give a >10× amplification of energy and a >4× amplification of the neutrons, as well as potentially destroying problematic actinide materials [91]. Reaction 5 shows a typical result of a $^{239}Pu$ fission with a fusion neutron. The example provided is for the highest probability fission product combination for a fusion neutron and $^{239}Pu \rightarrow {}^{104}Mo$ and $^{132}Te$ (JENDL 4 fission yield library). These nuclides decay quickly to $^{104}Ru$ and $^{132}Xe$. The energy yield accounts for energy lost to neutrino emission.

$$ {}^{239}_{94}Pu + {}^1_0n \rightarrow {}^{104}_{44}Ru + {}^{132}_{54}Xe + 4{}^1_0n + {\sim}190\ MeV \qquad (5)$$

The concept of such a "hybrid" fusion-fission system is not new, but it is technically immature so would require significant development [62] and is also dependent on the development of the associated fusion technology.

# Appendix 3: Safety and Environment Detriment

## Outline of SED Methodology

The radiological inventory is defined by its **Ingestion Toxic Potential** (ITP), which is the volume of water required to dilute a material to a concentration that would be safe to drink if a population were to use the mixture as its sole source of water. ITP is calculated separately for each radioisotope in the waste and summed. For isotopes and wastes for which dose by inhalation is likely, an additional factor, the **Aerial Correction Factor** (ACF) is added to the calculation. The effect of ACF can be considerable, with all plutonium isotopes having an ACF of around 480.

Chemical hazards are assessed by using their inventories compared to their COMAH (Control Of Major Accident Hazards) classifications as a measure of hazard potential, the **Chemical Hazard Potential** (CHP).

The ITP is then combined with a Form Factor (FF) which assesses how likely the waste is to disperse (high for liquids, medium for powders and sludges, low for discrete solids), and a Control Factor (CF) which assesses the stability of the waste – requiring revisiting after hours, days, years or decades. All these factors are combined as the **Radiological Hazard Potential** (RHP):

$$Radiological\ Hazard\ Potential = \frac{Inventory \times Form\ Factor}{Control\ Factor} \qquad (6)$$

In situations where the CHP of a substance is significant (for example the HF hazard presented by $UF_6$ reaction with moist air), then the CHP is calculated by a similar formula based on the COMAH limits of the particular chemical.

**Form Factor** (FF) is the form in which the material would be released if the containment was completely lost for approximately a day (e.g. for pyrophoric materials this would be a gas). It varies from 1 for gases and liquids (i.e. 100% release) via discrete solids at 0.00001 to large monolithic and activated components at 0.000001.

The physical, chemical and radiological properties of radioactive materials vary widely, and a factor is needed to reflect these attributes. The **Control Factor** (CF) addresses this, and considers "how difficult is it to store any given material?". This will take into account whether the material:

- Generates heat,
- Evolves flammable or explosive gases,
- Is corrosive,
- Is unstable (i.e. would degrade or react if exposed to air or water), or
- Would require additional systems to maintain its current physical state (e.g. agitation to keep solids in suspension).

All of these would affect the amount of human activity needed to control conditions during storage.

A reasonable yardstick by which to assess "difficulty of storage" is the length of time that the radioactive material could be left with no monitoring or other intervention, whilst retaining confidence that containment would be maintained and the material under scrutiny would remain controlled.

A long time here will indicate an easily stored material, while a short time will indicate one that is more difficult to store.

The RHP CF therefore attempts to take account of the differing intrinsic properties of radioactive materials by posing this question:

> *"For the current/proposed storage mode, what monitoring period would a competent and experienced design engineer be likely to propose at the design stage as being necessary to ensure containment, based on the intrinsic hazard of the material, the mode of storage, and how these might evolve?"*

Values of the CF are derived by engineering judgement. It relies upon judging the idealised radioactive material storage mode, rather than considering the actual condition of the current storage facilities. The CF reflects the monitoring interval, which ranges from hours to decades depending on the type of material and stores (e.g. open tanks, closed tanks, storage building or boxes). This is not the same as the actual frequency that would be applied in the design of a real plant operating within a valid justification of safety regulated by the ONR as, in real situations, continuous monitoring is often applied as a prudent measure.

## Safety and Environmental Detriment

The **Safety and Environmental Detriment** (SED) score for the waste is then calculated using the formula:

$$SED = (RHP + CHP) \times (FD \times WUD)^4 \qquad (7)$$

Here, the **Facility Descriptor** (FD) is a measure of the suitability of the waste store, and the **Waste Uncertainty Descriptor** (WUD) reflects the nature and condition of the waste: "Is it getting more difficult to deal with the longer it is left?", and "Will this significantly affect the approach to remediation?". Both of these factors have 10 categories attracting scores of between 2 (best) and 100 (worst).

The FD has defined scores between 2 and 100, with 100 scored by:

> *"Building past its original design life/intent, single containment, known/believed significant defects, and limited contingency provisions. Building is not qualified to withstand modern design basis hazards."*

...down to "stores to current standards" which score 2 and are described as:

> *"Building still within its original design life/intent, and no known/believed significant defects, has double containment, and well worked up contingency provisions. Inventory retrieval period would not extend beyond the remaining design life available. Building is also qualified to withstand modern design basis hazards and all project/facility safety case follow-up work is being implemented. Building not at risk from adjacent facilities, and doesn't itself present a risk to neighbouring higher hazard buildings/facilities."*

The WUD is similar, with a score of 100 ascribed to:

> *"Building contains raw wastes/by-products which are expected to be physically degrading with the potential for dissociation/dispersion of the material such as might affect retrieval methods needed, increase dose uptake for retrievals, or generate a criticality potential, and whose degradation is not being routinely monitored and managed."*

...whereas a score of 2 is given for:

> *"Building contains packaged wastes which are not reactive and known not to be physically degrading through monitoring/management programmes. Packages are qualified against design basis faults."*

A further element, which will not be further detailed here, is that there is also a SED-based methodology for assessing contaminated land.

## SED Scores of Plutonium Forms

As detailed above, all the new stores and stored wastes share the same CF, FD and WUD. There will be only two SED scores for the plutonium products which are the subject of this report – one for a FF of 0.1 (powder) and one for a FF of 0.00001 (discrete solid).

The major driver of hazard is the small particle size of the stored $PuO_2$. All the future options, both *Use* and *Dispose*, involve converting the $PuO_2$ powder into a solid form, so all future routes follow a path leading to large reductions of hazard. If stored as powder, there is only a small reduction in hazard during storage for periods of many decades. This means that all the paths which are being examined here have, once the materials are stored in state-of-the-art stores, only two SED values – one where the plutonium is stored as a powder, and one where it is stored as a solid. This would clearly change if the solid contained any additional ingredient with non-negligible CHP or RHP.

## Limitations of the SED Methodology

It is important to emphasise that a SED score is NOT an absolutely justified quantity – the formula was put together so that it produced results that seemed sensible after a rigorous examination by a stakeholder team with very broad representation (i.e. industry representatives, regulators, local nuclear site stakeholders and nuclear sceptics). The Prioritisation Working Group all agreed, after considering many examples, that indeed, "a bad score was worse, and a good score was better".

As explored above, the SED methodology was driven by the need to assess old wastes of uncertain properties and contents, stored in old stores and silos which were often in uncertain condition, and had never been designed to meet current engineering and safety standards.

When, in contrast, future disposition routes are considered, all the wasteforms and fuels generated would be highly stable and would be specifically matched to engineered stores meeting the most modern specifications. In these situations, the CF in the RHP would be likely to be years or decades, whereas the FD and WUD would each merit the best available score of 2, and any options sharing the same plutonium inventory and FF will be likely to share the same SED score.

A further limitation of the SED methodology is that it was designed to assess static situations where the materials are in-store. Clearly there could be deemed to be an instantaneous risk increase whenever a waste is retrieved, transported and/or conditioned, but the processes to be used will have been the subject of a rigorous safety case analysis and engineered to modern standards. It is this regime which ensures safety during processing and transport, and the SED has no role to play. Also, if more than one process is contemplated for the same activity – for example pond or cask storage – both will have been engineered and assessed to the same standards, and the SED score will apply equally to all processes.

Disposal in a GDF was also not considered in the SED methodology development. However, SED does provide a scoring method, as the disposed waste is committed to a repository meeting the GDF disposal safety case, with no intention to examine or re-treat it, essentially for infinite time. This infers that the CF becomes infinite, leading to a SED score for GDF-disposed wastes of 0.

## Malicious Releases

The SED measure was not developed to examine malicious releases of nuclear material (where materials are stolen from stores or in transit), or by disruptive attacks on the stores themselves. However, as discussed above, the FF, which makes up an important part of the RHP, varies from 1 for gases and liquids (i.e. 100% release) to large monolithic and activated components at 0.000001. It is reasonable to assume that this factor will be reflected in the release of material in the event of, for example, a dirty bomb, with material stored as a powder being 10,000× more contaminating than a discrete solid. While this is not directly relevant to malicious behaviour, it may have some utility in some areas.

## Hazards and Risks in Plutonium Management Options

The options for managing the plutonium inventory all start with the inventory "as is" – i.e. with a proportion in less-than-optimal containment and storage conditions and subject to an improvement programme – the extent and duration of which has already been mentioned.

With this programme assumed to be completed on the same timescale for all future management options, we can examine the SED formula (Equation 7). As previously stated, we are justified in assuming that any options sharing the same plutonium inventory and FF will be likely to share the same SED score, whereas the SED scores for a $PuO_2$ powder will be a factor of 10,000 higher than those for a solid fuel or wasteform.

It should be noted that the study shown in Figure 5 (page 35) was performed in 2017, on the assumption that the both the containers and the storage conditions were close to ideal. In fact, when the stock and its storage were systematically studied, it proved that neither the containers nor their storage conditions were actually ideal. This gave rise to the improvement programme which has been referred to and would have involved a considerably larger SED score than that shown in the figure (order of magnitude of $10^{22}$).

## SED as a Progress Measure

The exponential derivation of the SED parameter means that reductions in the amount of a stored plutonium species as activities (such as converting to solid from powder) progress show a relatively small reduction in SED until the conversion process is virtually complete. This is illustrated in Figure 6 (page 35). This is because a high hazard continues to exist, even with a relatively small amount of problematic material.

It follows from this that, at least as assessed by the SED methodology, the hazard reduction from converting stored $PuO_2$ powder to a solid form is similar whether this solid form is a wasteform or MOX fuel before irradiation.

For irradiated MOX fuel, the hazard as measured by SED will be very much larger than for the unirradiated fuel. However, the accessibility of the irradiated fuel is so much more difficult that hazards associated with malicious intent are highly unlikely to increase.

The key observation from the above is surely that the futures involving plutonium storage, whether followed by *Use* as fuel or *Disposal* as a wasteform, will involve similar reductions in hazard as assessed by SED from "improved containers and storage" to the onset of "*Use* or *Disposal* of the fuel or wasteform". This means that, once the current repackaging and storage in modern facilities is complete, the progress in hazard reduction over the next few decades will depend almost entirely on the timescales for changing the form of the $PuO_2$ by building and operating the plants to manufacture waste for disposal or fuel for irradiation.

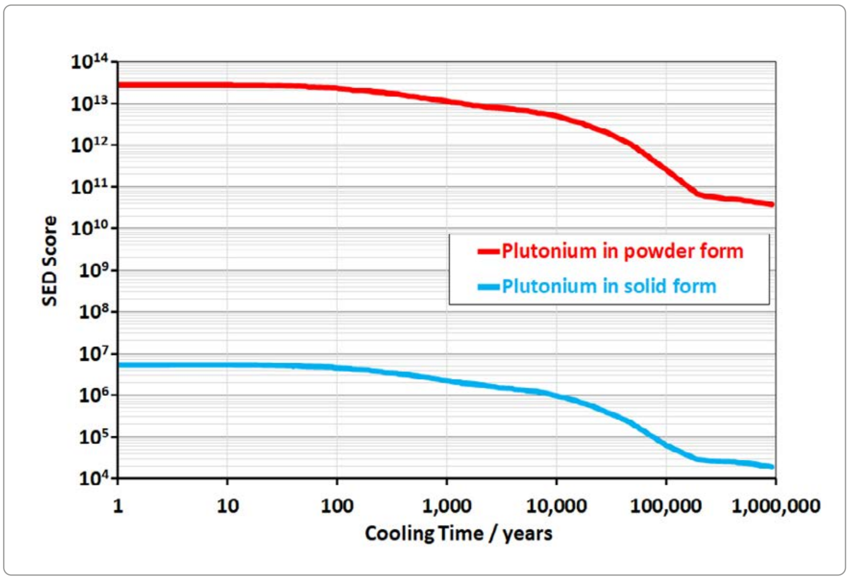

**Figure 5. SED score of $PuO_2$ wasteforms with time, assuming near-ideal storage conditions. Adapted from [63, Fig. 3].**

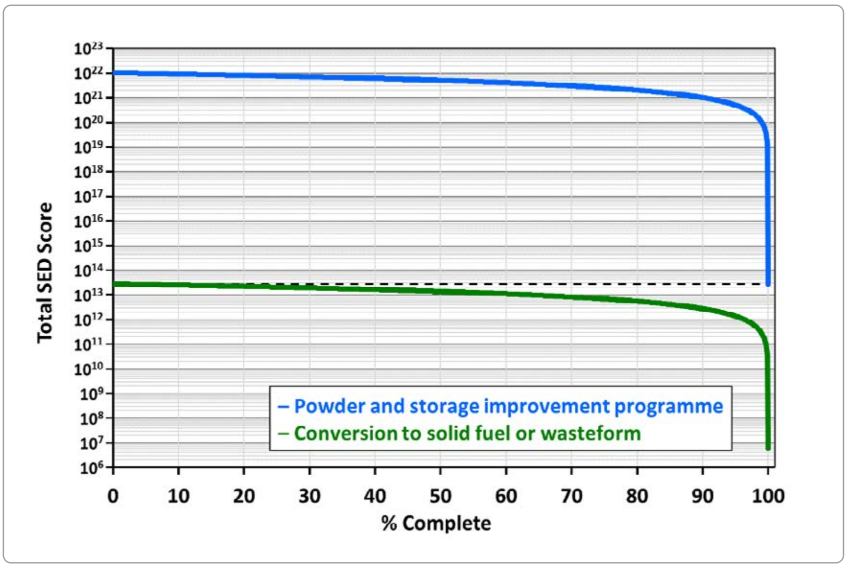

**Figure 6. Hazard evolution during inventory activities using SED measure.**

# Appendix 4: Background to Options and Assessments

## The Complexities of Plutonium Materials

Plutonium is a complicated element. It does not behave like any other element in existence because of unique electronic and nuclear properties which are confounded by its radioactivity. Its behaviours are difficult to predict, and difficult to control. Although it has been studied for fewer than 100 years, its use in nuclear technology has prompted extensive research into aspects of its behaviour. In the context of the UK stockpile, $PuO_2$ is the main material of interest although minor amounts of other materials such as plutonium carbide and plutonium chloride are also present. Materials such as zirconolite ($CaZrTi_2O_7$), manufactured as wasteforms, are also of interest [64]. Any plutonium material will be subject to a number of phenomena which make its characterisation, handling and storage, challenging.

## Radiation Damage

The decay of $\alpha$-emitting plutonium isotopes proceeds with the ejection of an $\alpha$ particle (a helium nucleus) and recoil of the decay product atom, which both cause radiation damage within the material. This self-irradiation leads to displacement of atoms in the crystal structure, with the $\alpha$ particle creating hundreds of atomic displacements and the recoil atom causing thousands of displacements along their paths. These damage cascades create vacancies (missing atoms) and interstitial atoms (atoms within the material that lie in-between the other rows of atoms) as well as extended defects in the material, resulting in lattice expansion of actinide compounds [65–67]. In some examples, including the fluorite-structure dioxides like $PuO_2$ [68], this expansion reaches a plateau, reflecting an ability of the structure to accommodate radiation damage [69, 70]. In this self-healing process, sinks such as grain boundaries, pores or crystalline defects within the material act as traps for interstitial atoms and vacancies, leading to annihilation of further produced defects and hence to quasi-steady state conditions in the bulk material [71]. The low barrier to activation for the recombination process allows it to operate effectively even at room temperature.

## Phase Change

All the $\alpha$-emitting plutonium isotopes decay to relatively long-lived uranium isotopes so uranium atom concentrations of up to ~2 at% will arise over a ~50 year storage period. Similarly, $^{241}$Pu decays to $^{241}$Am so americium concentrations of 2-3 at% will be present within a few years of separation and, over some decades, concentrations up to ~8 at% may arise.

$PuO_2$ has the cubic fluorite (F-type) structure, as do uranium dioxide and americium dioxide ($UO_2$ and $AmO_2$). While uranium, plutonium and americium are stable in oxidation state IV, americium also has relatively easy access to the +III oxidation state leading to $Am_2O_3$. $Am_2O_3$ has a complicated solid state chemistry [72–74], with the trigonal A-type, monoclinic B-type, and a body centred cubic C-type polymorph, related to the cubic structure of $AmO_2$, all known. The B-type structure is believed to require local stabilisation, and the C-type structure can be hyperstoichiometric in composition. AmO has also been reported. Formation of americium, followed by exsolution and segregation of an americium phase, could lead to localised distortion of the host fluorite structure.

Uranium also has access to oxidation states other than IV. In addition to $UO_2$; discrete $U_3O_7$ (tetragonal)/$U_4O_9$ (derived from fluorite structure) and $U_3O_8$ (layered, orthorhombic) phases – which contain mixed oxidation states – are also known, as well as $UO_3$ [75]. Certainly, in U/Am oxide systems, the americium is known to be present as Am(III) while uranium adopts oxidation states V/VI [76], and IV, V and VI [14]. Tracy et al. have demonstrated a relationship between radiation-induced redox changes and structural transformations in actinide materials [78].

## Compositional Change

Decay products are formed in $PuO_2$ atom-by-atom. Localised radiation damage, which is primarily caused by the recoiling product nucleus rather than the α particle, will initially leave the decay product uranium isotopes in damage cascade regions of the lattice, potentially leading to enhanced mobility. This could lead to chemically reactive surfaces or areas. By contrast, $^{241}Am$ is produced by $\beta^-$ decay and will thus not form in similarly damaged regions. The chemical and structural environments of the decay product nuclei are thus crucial to understanding aged $PuO_2$.

The presence of helium from α-decay within the structure can also fundamentally alter both the micro- and macroscopic properties of materials [79]. Importantly, complete disintegration of a 40 year old $PuO_2$ pellet was attributed to helium embrittlement [80]. Radiation damage facilitates helium mobility in $UO_2$ [81] and thermal desorption data reveal relatively complex behaviour in $(U,Pu)O_2$ [80, 82], while Tian *et al.* [83] have identified relationships between helium behaviour and $PuO_2$ composition.

Due to the complexities and the dynamic nature of the radioactive decay of plutonium isotopes, the UK stockpile, some of which has been stored for ~60 years, will have undergone significant alteration. To support the recommendation that the $PuO_2$ powder stockpile is converted into a densified form as soon as possible, it is imperative that the condition of the powder is understood and the effect on any manufacturing process can be removed, tolerated or mitigated for the vast range of parameters such as storage age, isotope content, and burnup. This will inform any pre-treatments that are required during the repackaging operations at SRP, predict the materials properties during that storage and allow the plant construction to accommodate the predictive models of material condition when it is retrieved for manufacturing into the final disposition form.

## Plutonium Compounds of Importance

### Plutonium Oxides

Plutonium forms a range of compositions with oxygen. UK civil plutonium is commonly found as $PuO_2$ (Pu(IV) oxide). $PuO_{2.00}$ is termed stoichiometric, where all sites in the crystal structure are occupied. However, because the material can accommodate vacancies particularly of the oxygen atoms, strictly, the material is non-stoichiometric, which means small amounts of oxygen are missing in defects – such as $PuO_{1.95}$. A range of oxides is known, including PuO, $Pu_2O_3$ ("sesquioxide" – two forms), $PuO_{1.61}$, $PuO_{2.00}$, $PuO_{2-x}$ and $PuO_{2+x}$ although the principal material of interest here is $PuO_{2.00}$. Consequences include changes in unit cell volume such that the crystal may shrink or increase in size, leading to high internal stresses potentially leading to cracking and changing the physical form and properties of the bulk material.

### $PuO_{2+x}$

There is agreement that plutonium can form molecular compounds with oxidation states V, VI, and VII, however, these tend to be formed in solution. There is disagreement on the existence of extended solids which incorporate more oxygen than $PuO_{2.00}$ in hyper-stoichiometric phases. That said, there have been reports of solids which correspond to hyper-stoichiometric phases such as $PuO_{2.26}$. Importantly, in the view of material storage, this has been reported to be formed from reactions on the surface of $PuO_{2-x}$ where $H_2O$ adsorbs strongly below 120°C, and desorbs as the temperature approaches 200°C. Dissociation of water forms reactive $OH^-$ and enables the formation of $PuO_{2+x}$, releasing hydrogen gas. Other mechanisms include surface adsorbed water that reacts with $PuO_{2-x}$ – $PuO_{2.00}$ forming $PuO_{2+x}$ and adsorbed atomic hydrogen on the surface in the presence of oxygen. The unit cell parameter for sub-stoichiometric $PuO_{2-x}$ is very sensitive to composition. However, once stoichiometry is reached in $PuO_2$, the unit cell parameter does not change much but this phase is of importance as little is known about its reactivity or long-term storage properties. Structurally, it is thought that excess oxygen atoms reside in the interstitial octahedral sites in the fluorite structure – charge balance, therefore, is thought to come from Pu(IV) becoming oxidised to Pu(V) or Pu(VI). The effects of the existence of these oxidation states in the presence of decay products, e.g. U(IV – VI), Np(IV – V) and Am(III – IV) are unknown and are important to investigate.

### Disposition Ceramics

Before any decisions are made regarding the immobilisation and disposition of plutonium, long-term and comprehensive research programmes are needed. Currently, it would be unwise to select a ceramic based on the current body of scientific and engineering knowledge and capability to support a decision for disposal in a GDF where wasteform stability on a 100,000 year timescale would be desirable.

## Fuel MOX

Reactor grade MOX for thermal nuclear power plants has been manufactured on an industrial scale globally using two routes; the SBR developed in the UK (discussed more in Appendix 1) and the MIMAS route, developed by Belgonucleaire in the 1980's and used in the French Melox plant. Thus, the technology is mature and deployed at scale, representing the most advanced TRL (Technology Readiness Level) for manufacturing of ceramics for disposition of the UK stockpile as reactor fuel. The two routes differ in their processing but broadly involve mixing $PuO_2$ and $UO_2$ powders before pressing into a pellet and sintering at high temperature. For thermal reactor MOX, about 7% $PuO_2$ is mixed with depleted $UO_2$ – in the SBR process these powders are directly mixed together, however, in the MIMAS route a master blend of around 30% $PuO_2$ is mixed first prior to down-blending with further $UO_2$ powder to achieve the final desired loading. This can lead to the formation of plutonium rich islands in the MOX fuel which may cause issues for reactor operation as well as reprocessing and their presence needs to be controlled through strict specifications. In addition to this, other fuel specifications such as pellet dimension, density, grain size and stoichiometry, may all affect the fuel's mechanical and thermal properties and must all be met to qualify for the fuel for use in reactor.

As discussed, the UK stockpile has spent varying lengths of time in storage where it has been exposed to self-radiation damage, decay product in-growth, adsorption of gaseous species and reaction with storage container materials. Further information in Appendix 1 illustrates possible methods of treating out-of-specification $PuO_2$ powders to reduce chlorine contamination, increase surface area and remove carbon impurities to bring that material back into specification. However, radiation damage defects, americium in-growth and potential helium gas bubble formation within the powder particles may also alter their manufacturability into a high-quality MOX fuel pellets. The effect of these parameters (which vary with $PuO_2$ storage time, reprocessing history, isotopic content) on pellet quality need to be understood. This will address the understanding of powder feedstock variability on pellet quality for current UK stockpile material. However, after the retreatment and repackaging program, these $PuO_2$ powders may be stored for several further decades before manufacture and so predictive modelling of how these variables affect pellet quality and plant operations would need to be understood as to design a suitable manufacturing process and plant for full-scale operations from 2050 onwards.

If the UK were to pursue a new MOX fuel plant, the first decision would be which route to base the plant on: SBR, MIMAS or a potential third route. With the closure of the SMP and cessation of the SBR along with the lengthy operational experience of the Melox plant and MIMAS process, it may be lower risk to adopt MIMAS for the UK. However, the UK's $UO_2$ powder feedstock differs from that used in the MIMAS process and so validation of UK $UO_2$ with the MIMAS route would be required. If the UK were to pursue a fast neutron reactor programme, fast reactor MOX would likely be the preferred candidate fuel form in the near-term. In fast reactor MOX, $PuO_2$ loadings are more typically ~20-30% but powder processing and pellet dimensions remain similar to thermal MOX fuel. Due to the higher burnups, the specifications for fast reactor fuel are different, requiring a higher degree of porosity for fission gas accumulation as well as different specification in grain size and plutonium homogeneity. However, the fuel specification and licensing process would need to be developed as part of any fast reactor programme and similar challenges to a thermal MOX programme would need to be addressed to ensure product quality if utilising the UK $PuO_2$ stockpile in this way.

## Zirconolite and Pyrochlore

Several candidates have been proposed for the immobilisation and disposal of plutonium, including crystalline ceramic materials as well as glasses. Zirconolites, a type of mixed-cation ceramic ($CaZrTi_2O_7$), and $Gd_2Ti_2O_7$ pyrochlore have been studied as candidates due to their natural occurrence and stability with up to 30 wt% uranium and thorium [84]. However, to test their long-term radiation stability with higher specific activity alpha-active actinides and support their use for disposal, several simulant methods are available. Accelerated damage studies use very high specific activity actinides or charged particle irradiation to simulate 100,000 years of damage on a laboratory experiment timescale, while study of natural analogues with U/Th is also useful. However, the rate of radiation damage varies wildly across these techniques which makes drawing conclusions and forming predictive models very difficult. Due to the ease of working with non-active analogues, most of this work has included the use of cerium and other surrogates for radioactive elements, as well as use of ion irradiation along with some research using natural or depleted uranium being reported. Accelerated damage studies with $^{238}$Pu [85] and $^{244}$Cm [86] doping have been performed which show the materials undergo significant amorphisation (loss of crystal structure) which results in significant swelling of up to 10 vol% and would occur prior to ground water breakthrough to the wasteform. However, the pyrochlore phase with composition $Gd_2Zr_2O_7$ has been shown to be dramatically more resistant to irradiation under $^{241}$Am doping and ion beams, remaining crystalline and not undergoing dramatic amorphisation swelling [87].

Although these minerals are found in nature, to form the desired phase with the correct structure type, the starting materials need to be very pure, and the process and synthesis conditions relatively strict. Undesirable side-products can form such as $ZrO_2$, $TiO_2$, $PuO_2$, perovskite or pyrochlore phases which have differing radiation resistance and swelling rates. These differences build up significant internal stresses and result in cracking which may accelerate leaching of radionuclides out of the wasteform. The addition of poisons such as $HfO_2$ or $Gd_2O_3$ to the material as neutron poisons to prevent criticality during disposal further complicate the chemistry, microstructure and potentially the mechanical stability. There is presently limited evidence of successful preparation of single-phase material by more modern techniques (such as HIP). Longer-term ageing effects, such as the accumulation of helium and formation of nanobubbles (which may affect mechanical durability of the wasteform due to processes such as embrittlement of grain boundaries) or volumetric swelling are not well understood. Research results to date are also inconclusive and require further understanding.

These phenomena, which are not understood comprehensively, demonstrate that concerted and long-term R&D programmes are necessary before making any decisions regarding any target disposition phases, including work with authentic materials (i.e. plutonium, americium, neptunium and uranium), and artificial ageing. Furthermore, although pilot-plant facilities for HIP manufacturing have been set up, these have not undergone plutonium active trials to demonstrate feasibility. Given the lower TRL of the ceramic wasteform, a scale-up demonstration facility would be required prior to a full scale plant.

Any phase or form which either becomes or is designed to be amorphous would be very challenging to analyse during an R&D programme or as part of material surveillance. This is because of the lack of long-range structural order and the difficulty they present in analysing using techniques available. That is to say, one portion of a sample will be different, by definition, to another from the same sample or batch. This needs careful consideration as phenomena which may occur over the different time scales of interest may be very difficult to measure, model, predict and control. Furthermore, wasteform and waste package design would need to accommodate the associated drastic swelling and long-term leaching experiments. There would be a need to demonstrate no deleterious effects of amoprhisation, such as cracking and failure of the wasteform. Wasteform and package design would need to accommodate swelling and potential leaching from primary containment. There is a need for demonstrable proof that any deleterious effects of amorphisation or change of the wasteform over time would not result in can failure and release of contents.

## Disposal MOX

Disposal MOX (also referred to as low specification MOX), is a candidate wasteform for the disposition of plutonium in a GDF due to its high radiation resistance and experience of industrial scale manufacture. However, unlike fuel MOX there is no current specification for disposal MOX and significant work will need to be undertaken to develop this to substantiate the safety case for geological disposal. Loadings of $PuO_2$ will be similar to, or slightly higher than for thermal MOX (around 10 wt% $PuO_2$), but as with the thermal MOX option, a decision on a route to industrial scale manufacture of a disposal MOX wasteform would be needed. In particular, a MIMAS, SBR or alternate route would require specification. Due to the differing challenges of developing safety cases for storage, transport and disposal (rather than licensing a fuel for use in reactor), the specification of $PuO_2$ loadings, pellet dimensions, porosity, grain size, plutonium-rich inclusion size distribution and the use of neutron poisons to prevent criticality during disposal are all currently unknowns which require investigation.

The initial design steps for the disposal wasteform would determine the MOX pellet dimensions. Without the restrictions of fuel assembly geometries, disposal MOX pellets could be much larger than fuel MOX pellets, with a diameter potentially >20 mm. This would reduce the number of pellets required to dispose of the stockpile as well as reduce their surface area to volume ratio to limit corrosion rates if ground water were to penetrate the wasteform. It is also possible that cross-sections other than cylindrical could be used, varying the packing density of the pellets. However, pellets this large and/or of different shape have not been manufactured at industrial scale so industrialisation is needed and practical limitations on achievable pellet dimensions against quality would need to be assessed.

The loading of $PuO_2$ would also require assessment and specification. A higher loading would reduce the wasteform volume produced, although there will likely be restrictions from criticality considerations. Optimisation based on parameters such as plutonium content, pellet size, shape and plant throughput will be needed.

Similar to the pyrochlore and zirconolite studies, accelerated ageing effects to mimic the self-radiation damage response and leaching behaviour would be required, and an understanding of the effects of microstructure (grain size and porosity) as well as dopant such as Gd(III) on the corrosion rates of a disposal MOX would all be required. Furthermore, if a neutron poison is required this would need to be accommodated in the design and implementation of the manufacturing process. For example, a MIMAS type route may require mixing poisons with the primary blend but the effects of this on plutonium-rich island formation, pellet quality and resultant performance would need thorough understanding to support disposal.

## Materials Characteristics of Importance

### Powder Morphology

Physical form of a solid is almost as important as its composition. The various methods used to produce $PuO_2$ and $UO_2$ powders give different crystallite and agglomerate sizes with a range of flow properties from free-flowing (e.g. $UO_2$ via the ammonium uranyl carbonate route) to "very not free flowing" (e.g. the UK Integrated Dry Route, IDR). This, when added to the variety of potential $PuO_2$ morphologies, leads to the requirement for a variety of pelleting and sintering routes and the potential for a range of quality problems. This means that an approach of "select the MIMAS route and away we go" misses out several levels of knowledge which will require significant investigation and proving.

### Minor Species and Contaminants

Radioactive decay generates product atoms (helium, uranium, neptunium or americium) in plutonium materials. $^{241}$Am, formed by decay of $^{241}$Pu (half-life 14.3 years), is particularly problematic. $^{241}$Am has a 433-year half-life and its $\alpha$-decay is accompanied by emission of 59.5 keV $\gamma$-rays. Thus, "aged" plutonium in which significant $^{241}$Am has formed presents an external radiation hazard which impacts on facility design, operation, maintenance and handling protocols. In addition to the radiological hazards, these decay products may also affect product quality which is discussed below.

### Americium

Americium can access II, III, IV, V, and VI oxidation states. Its solid-state chemistry is dominated by its III oxidation state, much like a lanthanide element, though three americium oxides are known, AmO, $Am_2O_3$, and $AmO_2$. The sesquioxide form, $Am_2O_3$, seems most likely since the presence of uranium in different oxidation states and plutonium – predominantly being in IV but with access to III, provide possible electron transfer pathways for the generation of Am(III) in situ. Formation of $Am_2O_3$ would present challenges in long-term storage and use of plutonium materials in powder feedstock since it has three polymorphs, all of which are structurally different from the host $PuO_2$ lattice. Exsolution and segregation of an Am phase could then lead to distortion of the overall bulk $PuO_2$ fluorite structure and hence to changes in parameters such as particle size/shape or packing. The behaviour and properties of solids containing plutonium, and significant quantities of uranium, neptunium and americium have not been adequately studied, and many phenomena which may be encountered would need significant R&D programmes to investigate.

### Uranium

Uranium can access the -I, I, II, III, IV, V, and VI oxidation states. Known solid-state oxide structures include a range of uranium oxidation states: $UO_2$ (IV), $U_2O_5$ (V), $UO_3$ (VI), $U_3O_8$ (the most stable U-oxide, disordered V and VI), $UO_2O_2$ (also known as $UO_4$, VI), and amorphous $U_2O_7$ (VI). The access to a wide variety of oxidation states in solids which have different structures raises the prospect of separation of uranium phases within the $PuO_2$ structure. However, the range of properties and stability of $UO_2$ during storage and processing with $PuO_2$ to MOX pellets is well known, so it is the change in properties during pellet storage both before and after inclusion in the GDF which is in need of considerable study. In fact, the attainment of this knowledge is likely to be on the critical path for satisfying the safety case for MOX pellet GDF disposal.

### Neptunium

Neptunium is a decay product in the decay chain of $^{241}$Pu via $^{241}$Am. It can also be produced by neutron capture in $^{238}$U. The principal isotope of interest is $^{237}$Np. Neptunium can access the III, IV, V, VI and VII oxidation states. In solids, the IV oxidation state dominates. As with plutonium, the Np-O binary phase diagram can accommodate defects and non-stoichiometry. However, there are only two known anhydrous oxide end members: $NpO_2$ and $Np_2O_5$. There is a large body of work to be completed to fully understand the

Np:O phase diagram [88]. Like $PuO_2$, $NpO_2$ crystallises in the cubic fluorite structure. $Np_2O_5$ crystallises in a monoclinic setting and is noted to be relatively unstable, decomposing to form $NpO_2$ and $O_2$ at temperatures between ~693 and 970 K.

### Poisons

Poisons are commonly used in fuel. In immobilisation and disposition, any phases nominated for further investigation would need their efficacy extensively tested in storage conditions, as well as accident and disposal conditions. That is, it must be easy to insert the poison into the structure of the host matrix* in a suitable concentration, must be in an appropriate chemical and radio-environment to perform as it should, and must not leach or form other compounds over time. This requires systematic and comprehensive R&D programmes using plutonium, uranium, americium and neptunium, since these systems are so complex that use of surrogates introduce major uncertainties.

### Multicomponent Systems

It is known that, at least in U-Am-O ternary systems, that uranium is present in V and VI oxidation states, while americium is present as III. The inclusion of plutonium makes the system much more complex and there are few studies of the Pu-U-Am-Np-O system [68], and particularly not on aged samples, either synthesised specifically or drawn from the stockpile. In the latter case, further complexity will arise from manufacturing variability and the presence of adventitious contaminants such as hydrogen, carbon, nitrogen or chlorine.

## Supporting the Assessments

### Research, Development and Innovation

At a high level, plutonium stewardship is simple. The material has to be stored safely and securely for some decades, then chemically and/or physically processed into a form which is suitable for whatever end-point is chosen by Government. All the steps along this path must be underpinned.

### Underpinning Storage

As described above plutonium materials will evolve over decades of storage. Since this evolved $PuO_2$ material will be the principal feedstock for whatever production process is required to manufacture the chosen end-product, the impact of evolution on material processability needs to be understood.

The integrity of storage also needs to be assured. Phenomena such as corrosion, degradation and pressure changes have been observed in previous generations of plutonium storage cans [89, 90] so understanding these processes, and monitoring their onset and progress, preferably without human intervention, will be necessary. The answers to these questions are central to the validity of the current SPRS programme of improvements to plutonium oxide condition packaging and storage.

### Underpinning Processing

While the specification, manufacture and performance through its lifecycle of fuel grade MOX is well understood, the equivalents for disposal MOX are not well defined, so this needs to be examined, especially as the lifecycle of disposal MOX is very different from that of fuel MOX. Moreover, the properties (e.g. specific surface area, contaminant content) of some stockpile material may be outside the specification for fuel MOX so the envelope for disposal MOX will need to be wider than for fuel material or the material will need to be reworked. Similar questions will need to be answered for other wasteforms such as zirconolite.

### Underpinning the End Point

As noted above, the disposability of irradiated MOX fuel has been explored extensively. However, the storage, transport and disposal of dedicated plutonium wasteforms is not yet defined. The nature and performance of the wasteforms and waste packages needs to be substantiated. The substantial increase in fissile inventory associated with disposal of plutonium as waste will need to be accommodated in the design and operation of the GDF.

### Tools and Techniques

There are parallels between the RD&I needs of the UK's plutonium management programme and the position of the US nuclear weapons programme around 30 years ago, where it was necessary to assure, over decades, the continued performance of the arsenal without full scale testing. This was achieved by "Science Based Stockpile Stewardship", the use of sophisticated research tools to develop predictive models of weapon performance. The UK plutonium management problem is technically much simpler but there would be great value in adopting the same approach.

Previously unavailable insights into the evolution of bulk $PuO_2$ and other relevant materials can now be explored using a range of techniques which are well established in

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* *The poison need not be added to the solid wasteform, i.e. not mixed within the plutonium-containing phase, but could be, for example, wrapped around a pellet as a foil, or present within the container material.*

other disciplines, for example the battery of synchrotron-based techniques, electron microscopies, or the X-ray pair distribution function. A wide range of techniques has been developed to characterise surfaces, which are so critical in controlling can environments and govern waste-form performance in the GDF. Data from these complex, challenging experiments, particularly so with plutonium materials, can support modelling to predict behaviour over a wide range of size and time scales.

A sustained commitment to RD&I underpinning the plutonium programme would be as transformative as it is essential. Developing and maintaining suitably managed subject matter experts – who are incentivised to stay within UK nuclear – will be vital. This includes more traditional, academic-trained National Framework Qualification Level 8 personnel, as well as more vocational routes toward similar expertise, knowledge and ability. This will limit the "leaky pipeline" of suitably qualified personnel, and ensure programmes are fully-furnished with continually developing expertise and who are properly managed and deployed where necessary input is needed. Broad engagement with UK universities with established and growing research centres for nuclear materials analyses is required, along with appropriately managed national facilities such as NNL and Diamond Light Source and AWE/MOD. The ARC Programme could be refocused on this integration task.

Knowledge transfer should be relatively simple to apply in a joined-up approach to solving the identified, and as yet, unidentified problems. Any R&D programme must have flexibility. This means that investigations into systems which would appear to be unsuitable following some investigation, are halted, and efforts redirected elsewhere. This will require materials scientists with a very broad knowledge of the solid-state and teams and facilities which are adaptable to sometimes rapid or unexpected change. This could occur at a fundamental research level, or plant-scale operations level to cope with the discovery of new knowledge, techniques, or specifications.

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