Research To Underpin The UK Plutonium Disposition Strategy

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ABSTRACT

In April 2005, the UK Nuclear Decommissioning Authority (NDA) took ownership of most of the civil nuclear liabilities and assets in the UK. These include separated civil plutonium stocks, which are expected to rise to over 100 tonnes.

Future UK national policy for disposition remains to be finalised. The feasibility of management options needs to be determined in order to allow the NDA to advise government on the ultimate disposition of this material.

Nexia Solutions has a contract with NDA to develop and carry out a research project which will result in a recommendation on the technical feasibility of a number of disposition options, focussing on re-use and immobilisation of plutonium as a waste for disposal. Initial work is already underway evaluating re-use with MOX and IMF fuels and immobilisation using ceramics, glasses and MOX for disposal.

The programme is expected to result, circa 2010, in a recommendation of a preferred route for immobilisation and a preferred route for re-use for the UK's civil Pu stocks.

INTRODUCTION

Until the early 1990's it had been intended to utilise the UK civil plutonium as fuel for fast reactors. However, in 1987, the Government decided to terminate the UK development programme. In 1994 the Prototype fast reactor (PFR) at Dounreay was shut down. As a consequence, there was no long-term strategy in place for the use or management of plutonium.

Separated plutonium stocks are held at Sellafield in the form of plutonium dioxide powder, using specially built stores that are subject to international safeguards and inspection by IAEA and Euratom. While plutonium has been stored safely and securely for many decades, storage itself is not a sustainable option and there remain concerns regarding proliferation resistance.

In 2005, the total holdings of UK-owned civil separated plutonium currently stand at around 70 tonnes. Based on anticipated arisings from reprocessing fuels from Magnox and AGR reactors the total could rise to about 100 tonnes over the next 10 years. At present the plutonium is regarded as a "zero value asset". Depending on future policy decisions the material may prove to be a major waste liability or a valuable energy asset.

In April 2005, the UK Nuclear Decommissioning Authority (NDA) took ownership of most of the civil nuclear liabilities and assets in the UK, which include these stocks. Currently future national policy for disposition remains to be finalised and the feasibility of future management options needs to be determined in order to allow the NDA to advise government on the ultimate disposition of this material.

Nexia Solutions has a contract with the NDA to develop and carry out a research project that will result in conclusions regarding the feasibility of a number of disposition options. These options have been developed from the recommendations of the innovative BNFL Stakeholder Dialogue process [1]. One key recommendation from that process was that plutonium disposition should be underway within 25 years (by 2025) and should be complete within 50 years (by 2050). Whilst these dates do not represent firm commitments, they represent major strategic targets on which the research project is based.

PROGRAMME OVERVIEW

As shown in **Fig. 1**, the three main options for the disposition of separated civil Pu are:

1. Continued storage - this will be necessary until agreement is reached on a long-term strategy
2. Re-use as fuel in existing or future reactors
3. Immobilisation in preparation for disposal

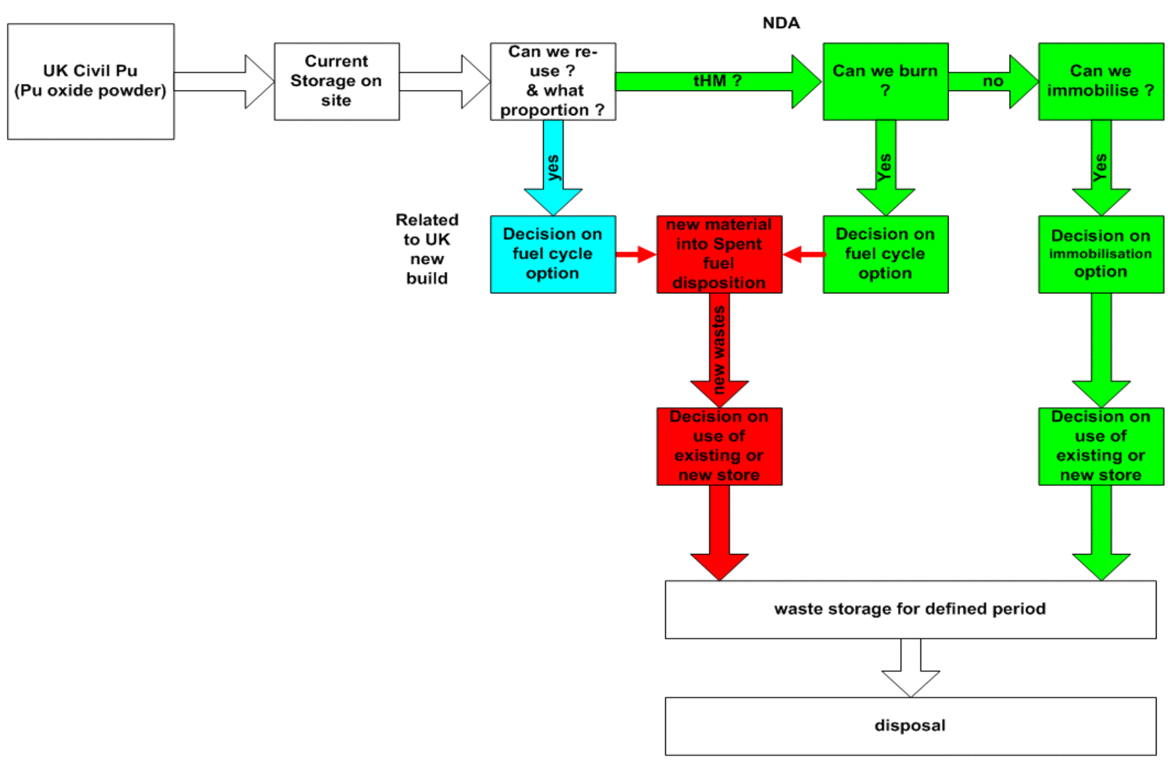

Fig. 1. Strategic option selection in the UK for civil Pu disposition

The decision, as to which option or options should be implemented, will be based on a consideration of a number of factors including technical feasibility, safety and environmental impact, political and stakeholder acceptance and economic viability.

The programme is divided into 3 areas of activity:

1. Strategic tasks – which support evaluation of technology options and provide information on safety and environmental impact, political and stakeholder acceptance and economic viability for the decision making process
2. Development of re-use options – which will provide information on the technical feasibility of candidate fuel cycles
3. Development of immobilisation options – which will provide information on the technical feasibility of candidate wasteforms

A phased approach has been adopted, with multi-discipline evaluations after each phase for selection of technology options. Fig. 2 and Fig. 3 show the phased development of re-use and immobilisation options. In general, phase 1 development will focus on a number of options on a broad front and with a limited amount of detail. Focus will be placed on those factors

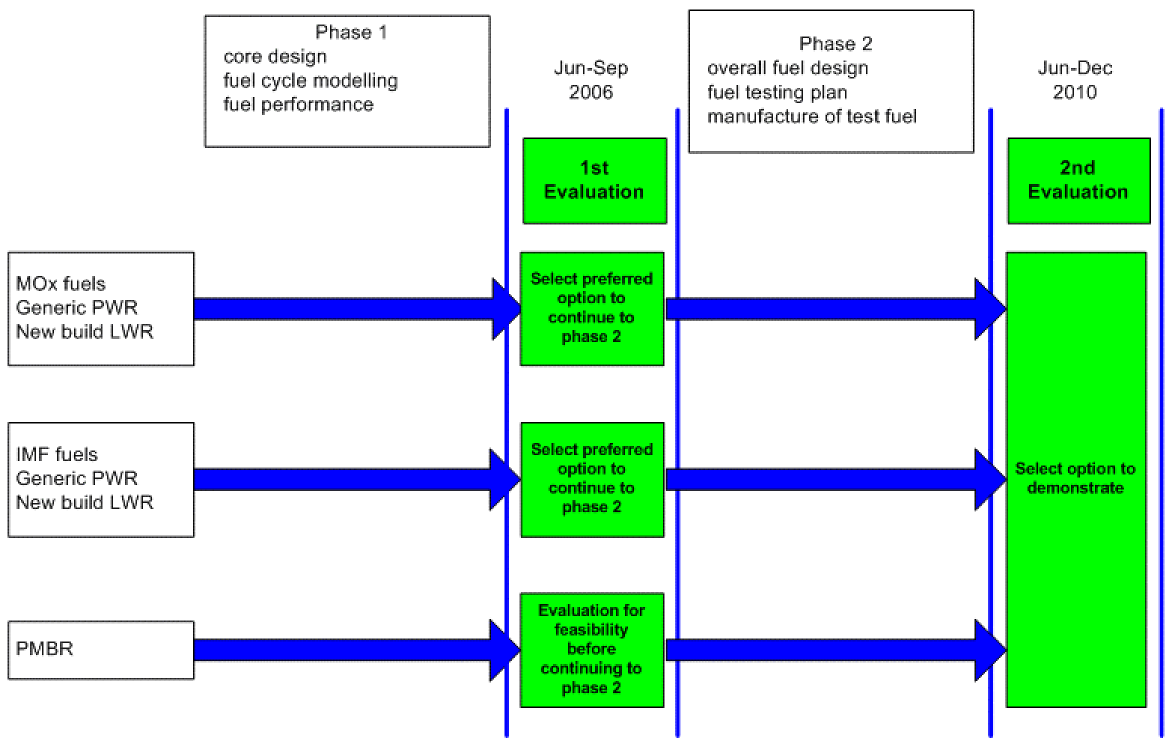

Fig. 2. Programme for Pu reuse

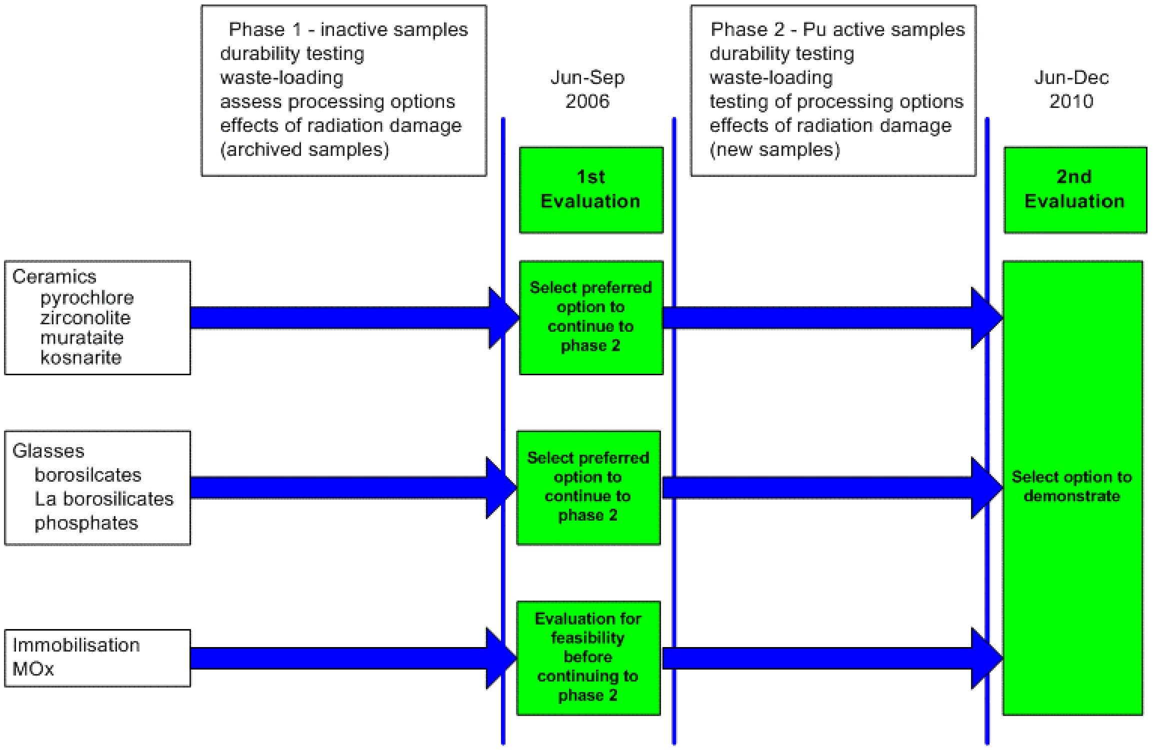

Fig. 3. Programme for Pu immobilisation

PROGRAMME FOR STRATEGIC TASKS

The main part of the strategic programme of tasks will be the evaluation of technologies. The activities are the key to the overall programme and will provide the basis of each stage of development. Each evaluation will be via a series of multi-discipline workshops. Firstly, a technical sub group will provide the initial evaluation for each technology group (e.g. immobilisation using ceramics). These subgroups will feed into a main evaluation taking into account multi criteria analysis of each likely scenario using the technologies. From this evaluation will result recommendations on which technologies are to be developed further.

The use of multi criteria analysis of implementation scenarios will require the development of a series of scenario analysis techniques to be applied to economic, environment and safety factors. Work has already commenced in this area and a series of indicators has been defined. A demonstration of the technique applied to a reduced set of environmental indicators for a selected set of Pu disposition scenarios will be completed at the end of March 2006. Nexia is using its participation in the EU 6th framework project, RED-Impact, to provide links to international practice.

A scheduling model will also be developed to assess the impact of implementation on UK sites and provide time-based data for the scenario analysis. A preliminary version of the model has been completed and will be providing initial data for the 1st evaluation workshop in 2006.

The final component to the strategic area is the generation of economic data. Cost estimation based of the implementation of each technology will be provided by a series of engineering design studies. These will be in phases and aligned with the R&D programmes, with initial studies provide “order of cost” data for the 1st evaluation workshop in 2006 and more detailed studies following on.

PROGRAMME FOR Pu RE-USE

In order to determine a research and development programme for plutonium re-use in UK reactors, it was necessary to assess the options available and then focus in on the strongest one. The UK expertise in a range of reactor and fuel cycle variants was key in being able to make such an assessment and necessary judgements in shaping the final programme. The expertise includes decades of MOX fuel manufacturing experience (fast and thermal reactors), design and licensing of MOX for utilities, fundamental understanding of reactor physics and fuel performance for UO_2 and Pu disposition as well as knowledge of safeguards. It is the integral capability and understanding within Nexia Solutions on how each of these key components impacts on all of the others that ensures that the final re-use programme achieves the end objective. Internationally recognised and state of the art tools either developed within or purchased by Nexia Solutions for the core assessments are then applied to ensure that the results are as accurate and relevant as they can be.

Plutonium fuel is in principle usable in the UK’s existing reactors and in any future reactors that might constitute a new build programme. However, each option will have its own strengths and weaknesses and therefore in order for a full assessment to take place, all of the options have been considered that are possible for both existing and new build reactors. For existing UK reactors, there are obvious limitations (such as the limited remaining lifetimes for Magnox, to a lesser extent, for the Advanced Gas cooled Reactors (AGRs)) that rule out any realistic prospect of making a significant impact on the stored plutonium. Nevertheless, even these have been considered for the sake of completeness. New build reactors have a much greater potential for plutonium irradiation and there are many possibilities which could satisfy a requirement to reduce the UK’s separated plutonium stockpile to close to zero within a few decades.

It was helpful to categorise the different options into four broad categories that were:

- Current reactors
- Near term reactor options (first operating by 2015)
- Medium term reactor options (first operating between 2015 and 2030)
- Long term reactor options (first operating after 2030)

All the UK’s commercial power reactors, comprising Magnox, AGR and the Sizewell B PWR are cover under current reactors. In principle, any of the current reactors could make use of plutonium fuel and they were all considered for completeness.

Near term future reactor options include all systems that are offered at present by the reactor vendors for which construction could start before 2015. Candidates include

- a replica of Sizewell B
- Westinghouse AP-600/AP-1000
- Westinghouse System 80+
- European Pressurised Water Reactor (EPR)

- ABWR
- Advanced Candu Reactor (ACR).

On the grounds that the UK has experience of PWR operation from Sizewell B, and no experience of BWRs, ABWR was excluded from the analysis. It was considered that there are sufficient generic similarities that BWRs can be considered encompassed within the PWR evaluations. On the grounds that British Energy have recently worked closely with AECL to assess the potential of the Advanced Candu Reactor (ACR) in the UK (as well as AP1000 in collaboration with BNFL and Westinghouse), ACR is also included.

Under medium term options are reactor systems which could be available in the next 10 years or so, but which are not available immediately. Under this category are PBMR (Pebble Bed Modular Reactor) and IRIS (an evolutionary design of PWR with an integral pressure vessel designed for passive safety). The medium term systems are assumed to become available in the timescale 2015 to 2030.

Categorised under long term options are four of the six Gen IV systems, SFR, GFR, LFR, and VHTR (Sodium Fast Reactor, Gas Fast Reactor, Lead Fast Reactor and Very High Temperature Reactor). Two of the Gen IV systems, MSR and SCWR (Molten Salt Reactor and Super-Critical Water Reactor) have been excluded on the grounds that both are of less interest to the UK. Moreover, in many respects SFR and LFR are very similar, so for the purposes of this review they were captured together under the heading Liquid Metal Fast Reactor (LMFR). Realistically none of the Gen IV systems will be available for deployment in the UK before 2030 or thereabouts.

The assessment undertaken as part of this study compared the relative merits of the different reactor systems and options in the context of irradiation of the UK’s plutonium stocks. The comparison was intended to be a high level one that avoided excessive detail; in order to highlight the relative advantages and disadvantages of the different systems and the common constraints that apply to all of them. This approach then identified the gaps in the existing knowledge base and helped form the proposals for the development programme required to fill these gaps.

Table I summarises the criteria and the key issues that were used in the assessment of the relative merits of the various reactors.

For brevity, it is not possible to present all of the detailed findings in this paper as the summary assessment equates to a report of more than sixty pages [2]. However, by way of example, one of the findings was that, although the use of IMF would be a viable option given the timescales, there is limited fuel performance knowledge and the modelling capabilities are limited within the existing fuel performance code. In addition, some of the fundamental reactor physics responses have not been investigated in detail in the UK. Therefore, it was recommended that the programme of work included a literature review of the fuel performance of IMF in order to improve the capabilities within ENIGMA-B and then undertake a comparison with existing test reactor data that Nexia have access to. This should also be complemented with a fundamental assessment of the reactor physics of IMF.

Overall, it was concluded that Sizewell B is the only current UK plant for which a plutonium irradiation programme would be favourable though it would be useful only for demonstrating early commitment to the eventual goal of removing the entire UK plutonium stockpile from immediate use. In the near term, new build based on evolutionary Pressurised Water Reactors (PWRs), such as AP-1000 or EPR would create the opportunity for the timely consumption of the entire UK stockpile within 20 to 30 years of the first new plant being commissioned. Other options based on new advanced reactor designs are expected to become available in the medium and long term such as with Generation IV. All these are capable of meeting UK plutonium mission goals, in many instances with some advantages over evolutionary PWRs e.g. sustainability. However, the advantages are generally not so large to justify delaying progress and the most sensible strategy would be to press ahead with plutonium irradiation in the near term reactor options whenever these are built.

Table I. Criteria Against Which Each Reactor Option Were Evaluated

Category	**Key Issues**
1. Timing issues	- Acceptable - Too quick - Too slow
2. Throughputs	- Are they known? - Sufficient to use all UK Pu stockpile? - Demands on fuel manufacturing
3. Pu Destruction rate	- High - Medium - Low
4. Fuel Options	- Standard/known - Risks? - Advanced options available?
5. Technical Viability	- High - Medium - Low
6. Strategic Flexibility	- Closes off future options? - How the option affects the future accessibility of the plutonium for a programme of new reactors, such as Generation IV demands
7. Safety	- Reactor - Fuel manufacturing - Transport
8. Proliferation Resistance	- Relative within the Reactor systems options
9. Waste Forms	- Known levels? - Known quantities? - Effect on radiotoxicity

The expert panel identified several areas for further work and this has formed the basis of the work programme now underway. The first phase of the assessment will focus on the option for Pu re-use in a generic PWR and will consider a fuel and core safety analysis using MOX and inert matrix fuels.

Evaluation Of Re-Use In A PWR

Previous studies on MOX in PWRs have been carried out in the UK, but this has primarily been in support of BNFL's MOX business or support to European utilities; rather than evaluate the Pu re-use options for application in the UK context. Furthermore, although it is clear that there is now extensive worldwide experience with MOX in PWRs, this experience is related to a plutonium vector that differs from that arising from the reprocessing of the UK Magnox and AGR fuels for example. Therefore, the principal aim of the first phase of this work is to complete an analysis that is specifically related to the UK context (licensing, plutonium vector, operating cycle lengths, mission of plutonium re-use etc) and therefore filling the gaps in the knowledge base with regard to core and fuel performance of a UK PWR using UK owned civil plutonium. This allows a true assessment of the plutonium burning potential of a generic UK PWR, based on existing PWR technology (rather than advanced plants such as AP1000) as well as identifying any key technical show stoppers associated with the fuel or core design. By completing such analyses, it also allows the UK to prepare the ground as much as possible by studies of specific scenarios that can be used as "off-the-shelf" references for any future detailed optioneering. These will be valuable in reducing uncertainties and make use of various sophisticated tools that are already available. At the very least, such studies will allow NDA to respond more quickly and from a more informed position from the perspective of an intelligent custodian of the UK owned civil plutonium.

The study will include a fuel management, reactor physics and fuel performance analysis in order to determine the feasible plutonium loadings. This analysis will then form the basis of a fuel cycle modelling exercise to look at the impact of the option on the civil plutonium stockpile and on the radiotoxicity of the proposed fuel cycle compared with a standard uranium dioxide (UO_2) fuel cycle and provide a benchmark against which to compare more advanced options.

An extension to this analysis will also assess the feasibility of using Inert Matrix Fuel (IMF) in a generic UK PWR. As in the case of the standard MOX analysis outlined above, a fuel management, reactor physics and fuel performance analysis will be completed to determine the feasible plutonium loadings. Again, this will form the basis of a fuel cycle model that will allow a comparison of an IMF fuel cycle with that of standard MOX and UO_2. In order to complete the IMF analysis, development work is required to modify the fuel performance analysis tool (ENIGMA-B) to allow IMF to be modelled more accurately.

PROGRAMME FOR PLUTONIUM IMMOBILISATION

One of the conclusions from the BNFL Stakeholder Dialogue process [1] was that a significant programme of work would be required to underpin the immobilisation of plutonium stocks, should immobilisation be the strategy chosen for the UK civil plutonium. This conclusion arose largely from the fact that little or no previous work has been carried out in UK, since hitherto plutonium has been considered an energy asset.

Researchers have been active in this area world-wide including a significant level of R&D that has been carried out for the US DoE to support the ex-weapons plutonium disposition programme, the immobilisation aspect of which is currently suspended.

The UK programme is aimed at providing options for the long-term storage and subsequent repository disposal of the UK civil plutonium stocks. These include plutonium derived from reprocessing of Magnox reactor fuel, which contains largely Pu-239, as well as AGR derived plutonium which have increasing levels of Pu-241, bringing with it the need for additional shielding. The design of any repository has yet to be defined and therefore the wasteform development must be carried out in such a way as to develop a product that will be suitable for disposal over a range of credible scenarios.

It is important to note that the plutonium for which these options are being considered is civil in origin and as such will be subject to the necessary safeguards both during processing, storage and disposal.

A review of previous research and development has been carried out with a view to compiling a programme of work that will explore a range of immobilisation technologies appropriate for UK plutonium stocks.

A broad-based approach has been outlined in which a number of different options will be studied leading to an initial and possible subsequent assessments aimed at eliminating the less suitable options. This down selection will allow the more in depth studies to be carried out on the one or more options thought most appropriate to UK needs.

The major options identified fall into 3 major categories: ceramics, vitrification and MOX as a wasteform, described below.

The project will use decision-making criteria which will facilitate such down selection and cover both product and process issues. As part of that a wasteform acceptance specification will be developed and agreement will be sought on such a specification from stakeholders.

The absence of a defined repository suitable to accommodate large quantities of plutonium containing wastes presents a challenge to the implementation of a wasteform specification. It is also accepted that the development of wasteform acceptance criteria will require significant buy in from UK stakeholders such as NIREX, EA and NII. In order to develop a wasteform it is necessary to assume some wasteform acceptance criteria in order to ensure that the best practicable options are chosen.

Ceramics

A literature review [3], supported by work being carried out on natural analogues under a project initiated by BNFL at University of Cambridge has identified a number of mineral phases that may be suitable for the immobilisation of plutonium. These range from pyrochlore, as favoured by the US ex-weapons plutonium immobilisation project, through zirconolite, a constituent of Synroc, to phosphate based phases such as kosnarite and murataite. The initial programme will focus on synthesis of these materials using cerium as a plutonium simulant. This will be followed in the later stages by active work with plutonium to validate the earlier results followed by in depth studies on the preferred options.

The ability of each candidate host phase to accommodate the plutonium is being assessed, using simulants. This will extend to the co-immobilisation of plutonium with associated neutron poisons aimed at preventing any criticality in the repository and following any leaching of plutonium from the host phase. Capacity for each of the phases to accommodate plutonium may be critical in assessing the cost effectiveness of the solution and as such waste loading limits for each candidate phase will be tested. Current work is underway at Nexia solutions and ISL (Immobilisation Science Laboratory, University of Sheffield) examining the formation and characterisation of pyrochlore and zirconolite crystal phases using cerium as a simulant.

Leach testing of Pu active samples will be employed in order determine a measure of durability as well as establishing the co-leaching of the plutonium with its associated neutron poison, critical to ensuring repository safety. Several techniques could be used and early in the program the appropriate technique will be selected.

Fabrication routes will be developed for the preferred host phases that can be demonstrated to be criticality safe as well as minimising dose to the operators and secondary effluents. These will be based on known technologies such as cold press and sinter and hot isostatic pressing.

In assessing host phases it is critical to understand their ability to accommodate a degree of radiation damage without over extended time periods without the possibility of changes to performance particularly when related to plutonium leach rate. The early fabrication of active samples using Pu-238 is planned giving time for radiation damage to occur and thus allow an assessment of that damage which can be taken into account when discarding the least preferable options.

Vitrification

Vitrification is an accepted technology for the immobilisation of fission product wastes resulting from the reprocessing of spent fuel. Considerable experience has been built up at Sellafield and elsewhere in the operation of such technology and vitrification is being considered as a possible route for the immobilisation of plutonium.

A considerable amount of data on composition and behaviour of a number of candidate glasses is available in the literature and we are building upon that data to develop a host matrix. The programme to assess these compositions is underway and in similar fashion to the ceramic programme may see a number of options quickly narrowed down as resulted of comparisons of wasteform performance. Borosilicate glasses similar to those used for fission product immobilisation, lanthanide borosilicate and phosphate compositions are being considered.

A major issue with the fabrication of plutonium glasses will be fabrication, as traditional melting routes may present criticality problems. For example the use of AVH (Atelier Vitrification La Hague) melter system such as that employed in Windscale Vitrification, Plant at Sellafield, can result in the settling of heavy metals in the melter heel under certain operational conditions. Some compositions chosen may also require higher melting temperatures that those traditionally employed during the fabrication of fission product glasses and effluent and handling issues may need to be addressed prior to them being considered as possible solutions.

Disposal MOx

A recommendation of the stakeholder dialogue plutonium-working group was that the use of MOx fuel as a wasteform should be assessed. Given that MOx fabrication technology has been employed on the Sellafield site, it maybe considered that production issues have been addressed and that MOx fuel with relaxed dimensional tolerances, not required for wasteform purposes, could provide an adequate wasteform for storage and disposal. A necessary first stage in this assessment is to ascertain durability of unirradiated MOx pins and comparison to durability behaviour of proposed ceramic and vitreous forms will give an early indication as to whether MOx as a wasteform could feasibly be considered for plutonium disposition.

Advantage has been taken of an existing agreement with ITU Karlsruhe who are already involved in an assessment of BNFL's irradiated MOx fuel as regards it possible future performance in a disposal scenario. This programme has been extended to include pellets of unirradiated MOx as they would be under a possible plutonium disposition scenario. Early results are expected in spring 2006 and will form the basis of a decision as to whether the material can be considered as a wasteform and thus be worthy of further investigation.

CONCLUSIONS

The NDA has commissioned Nexia Solutions to carry out a research project, which will result in conclusions regarding the feasibility of a number of disposition options for the UK-owned civil plutonium stockpile. This project is now underway.

The project focuses on 3 areas of activity:

1. Strategic tasks
2. Development of re-use options
3. Development of immobilisation options

Initial work on re-use options has identified the use of MOX and IMF fuels in evolutionary PWRs (e.g. EPR and AP-1000) as candidates for further study and work is now underway to analyse the fuel cycles in more detail.

After an initial review of options for immobilisation, work has begun on the assessment of ceramics, glasses and MOX as candidate wasteforms.

The phased approach adopted for the project will result in an evaluation of both re-use and immobilisation options, with a view to selection of preferred candidates for further and more detailed study. Currently, phase 1 of the project is scheduled for June to September 2006.

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