

Options For The Disposition Of UK Civil Plutonium Stocks

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ABSTRACT

The UK Civil Pu Disposition project will identify and assess the credible technical options for dealing with the UK's Pu stockpile and the discriminators that will support technology option down-selection decisions in the future. The Nuclear Decommissioning Authority's (NDA) role is in advising Government on the available alternative options and the consequences of potential decisions. The National Nuclear Laboratory (NNL) is supporting the NDA in this role.

This article briefly describes the work completed to date to assess the immobilisation and re-use technologies as disposition strategies and on the work that will help to identify discriminators to support eventual technology down-selection. The main focus of this article will be on work undertaken to investigate the possible criticality behaviour of the Pu stockpile in repository for different encapsulation and disposal methods. The Pu disposition project is anticipated to continue for several years while the necessary investigations are made to fully understand the various options.

1 INTRODUCTION

Upon completion of the currently scheduled reprocessing programmes, the UK Nuclear Decommissioning Authority will own a significant quantity of separated Pu. The disposition of this material is a key decision facing the UK Government. The NDA has an important role in advising Government on the available alternatives and the consequences of potential decisions. The National Nuclear Laboratory is supporting the NDA in this role, through the Civil Pu Disposition project.

There are three general strategic options available to the NDA for the disposition of Pu:

- Immobilisation as a precursor to long term storage or disposal
- Re-use as fuel in suitable reactor systems
- Long term storage – a deferment option which may not lead directly to disposal

The Civil Pu Disposition project will identify and assess several credible technical options for dealing with the UK's Pu stockpile and the discriminators that will help support technology selection decisions which may be based on several factors. The overall aim will be to deliver feasible technical options and information underpinning each strategic option.

Currently the first stage of the Civil Pu Disposition project is assessing a large number of technology options (both in re-use and immobilisation) at a broad level of detail. The focus is on establishing technical feasibility, identifying and addressing knowledge gaps and establishing process economics. The second stage will offer additional information on several credible technology options which may be used to inform the selection process at a later stage in the programme of research within the Pu Disposition project.

A brief review of the work completed to-date, to assess possible re-use and immobilisation technologies as disposition strategies is presented. The article then focuses on work undertaken to investigate the possible criticality behaviour of the Pu stockpile in repository for different encapsulation and disposal methods

2 OPTIONS FOR RE-USE

In order to determine a research and development programme for Pu re-use in UK reactors, it was first necessary to assess the options available and then focus in on the strongest one(s). Internationally recognised and state of the art tools, some of which were developed within the National Nuclear Laboratory, for the core physics, fuel performance and

fuel cycle assessments are then applied to ensure that the results are as accurate and relevant as they can be. The work presented in this article focuses on a possible re-use strategy within the UK, however re-use overseas may also be considered as credible re-use options.

The key overall aim of the re-use programme is to determine whether Pu disposition in reactor(s) can be achieved within the current UK design limits, whilst staying within international experience wherever possible. To answer this, the following factors were targeted and considered in establishing the individual tasks:

- Provide an informed position regarding the re-use options available to the UK which includes:
 - Which reactors are technically feasible?
 - What are the knowledge gaps?
 - What are the challenges/issues to be addressed?
 - Are there any “timing” issues? Eg. Pu ageing, facilities?
- Provide information to the “intelligent custodian” (i.e. the NDA) on the technical feasibility of candidate fuel cycles

These points are being addressed by undertaking work programmes that can be categorised into 3 main headings:

- Assessment of an existing UK PWR; Sizewell ‘B’
- Assessment of future UK reactor options; Westinghouse AP1000 [1] and the European Pressurized Reactor (EPR)
- Assessing advanced fuels (Inert Matrix Fuels (IMF)) for deployment in the above two reactor systems

It should be noted that in the case of the MOX evaluations, Pu sources from both UK Magnox and UK Advanced Gas-cooled Reactors (AGRs) were analysed.

Other options based on new advanced reactor designs are expected to become available in the medium and long term such as with Generation IV and the potential use of fast reactors. All of these are capable of meeting UK Pu disposition mission goals, in many instances with some advantages over evolutionary PWRs (eg. In the area of sustainability).

3 OPTIONS FOR IMMOBILISATION

Ceramic, vitreous and immobilisation MOX wasteforms (which is defined in a subsequent section) along with the possibility of cementation have been identified as the most appropriate for study as Pu host matrices, in phase 1 of the project. The wasteforms developed will require, in no particular order of importance:

- A demonstrated proliferation resistance
- Reduced likelihood of post-disposal criticality
- An established level of durability suitable for repository disposal
- Tolerance for high levels of radiation damage
- Economic waste loading
- Straightforward processing
- Chemical flexibility to accommodate impurities

It is important that the UK programme on Pu disposition is targeted at UK needs and requirements while taking account of the technical efforts that have been carried out elsewhere. To that end the following programme is being pursued.

3.1. Ceramic Wasteforms

A systematic study of ceramic phases suitable for Pu immobilisation has been carried out. Initial work on the examination of phase development and waste loading was carried out using cerium as a Pu surrogate. This has now been extended to the use of uranium. Several ceramic phases have been investigated and a range of phases have been shown to have adequate capacity to accommodate PuO₂ and neutron poisons such as Gd and Hf. Waste loading levels > 10% are shown to be possible and work is being carried out to evaluate higher loadings. A summary of potential ceramic wasteforms is shown in Table 1.

Table 1. Details of various ceramics under investigation for potential use as a Pu bearing wasteform

Phase	Structure ¹	Loading w% ²		Incorporate ³ Poisons	Sinter Temperature °C	Density % theoretical
		Ce	U			
Zirconolite	2M	<14	<13	Hf & Gd	1450	98
	4M	<33	tbd ⁴	Hf & Gd	1450	98
Zirconolite Ca substitution	2M	<26	<24	Hf & Gd	1450	98
	3O	<43	tbd	Hf & Gd	1450	98
Zr Pyrochlore	Pyrochlore	<19	tbd	Hf & Gd	1650	94
	Fluorite	<54	tbd	Hf & Gd	1650	94
Britholite		<67	<14	Gd only	1400	93
Kosnarite		<9	<18	Hf part Gd	1350	91

3.2. Glass Wasteforms

A wide range of glass compositions have also been fabricated and assessed for their suitability for the immobilisation of separated Pu. It has been demonstrated that Pu-vitrification is feasible and waste loading for surrogate materials of greater than 10 wt% in a number of silicate and phosphate glass matrices is practicable.

Following the initial evaluation of ten candidate glass systems, several compositions were selected to be taken forward for a more detailed assessment:

- *Modified-MW (MMW)* – the addition of a mixture of Al₂O₃, CeO₂, Gd₂O₃ and HfO₂ to MW (see below)
- *Lanthanum Borosilicate (LaBS)*
- *Alkali Tin Silicate (ATS)*
- *Sodium aluminium phosphate (NAP)*
- *Iron phosphate (IP)*.

Poor durability, inferior waste loading, evidence of crystallisation, and extremely high processing temperatures were among the factors that contributed to the rejection of the following compositions:

- Simple alkali borosilicate (MW)
- Calcium borosilicate (CaBS)
- Aluminium borosilicate (ABS)
- Lanthanum aluminium silicate (LAS)
- Lead iron phosphate (LIP)

The development programme has sought to maximise waste loading while at the same time reduce melt temperatures in order to facilitate ease of manufacturing. Initial melts were carried out using cerium as a Pu surrogate. While cerium has given a good indication of Pu solubility in glass, it has a tendency to autoreduce at high temperatures and as such is not typical of the Pu⁴⁺ state. As such, cerium has been replaced by hafnium as a surrogate which has the additional advantage of having a similar density to Pu and therefore has a more representative behaviour in glass melts where settling can be a major problem. This work has been completed for both ATS and LaBS glasses, but remains to be confirmed on MMW glasses without any Ce and containing solely Hf as the Pu-surrogate. A summary is presented in Table 2.

Table 2. Details of various glasses under investigation for potential use as a Pu bearing wasteform

Glass	Surrogate type	Surrogate loading w%	Melt temperature °C	Comments
MMW	Ce & Hf (Gd as poison)	>10	1300	Requires validation using Hf as surrogate
ATS	Hf	>10	1200	Viscosity at 1200 °C gives

¹ Different structures for the given material type

² The waste loading weight % which can be incorporated into the matrix material.

³ The neutron poisons which can be incorporated into the matrix.

⁴ To be determined

				pouring problems
LaBS	Hf	10	1400	Durability drop off at low pH

3.3. Unirradiated MOX Wasteforms

Another option for Pu disposition is the utilisation of unirradiated MOX pellets as a wasteform (immobilisation MOX) which would subsequently be disposed in a geological repository. To test the suitability of MOX pellets as a wasteform, samples with varying Pu content have been sent to the Institute for Transuranium Elements (ITU) in Karlsruhe, Germany, for leach testing. The pellets have been cut, polished to 0.25 µm and a surface characterisation has been carried out.

Short and long term static leach tests have been carried out at ambient temperature. These tests have been carried out with a range of leaching media; pure water, granitic water and carbonated water. Initial leach rates have been obtained from analysis of the short term tests. Results obtained to date show higher normalised leach rates observed for uranium than for Pu. When full analyses become available these results will be compared to those obtained under similar test scenarios on ceramic and glass wasteforms.

3.4. Cemented Wasteforms

A desk study is examining the viability of immobilising a stockpile of PuO₂ powder in hydraulic cement. The study has involved undertaking a series of calculations to predict the maximum loading of PuO₂ powder which could be incorporated into conventional intermediate level waste cement monoliths manufactured at Sellafield. These waste monoliths include 500 litre and 3m³ cement monoliths made from hydraulic cement consisting of formulations of ordinary Portland cement blended with blast furnace slag or pulverised fuel ash. The calculations compare the internal gas pressure developed inside each monolith from the alpha radiolysis of cement pore water with the strength of the cement at a given age to assess maximum PuO₂ loadings. This work is on-going.

3.5. Criticality Modelling

One of the key influences on the design of Pu containing wasteforms is the ability to mitigate against criticality in both storage and disposal scenarios. Various modes of criticality have been previously identified by US researchers and the use of neutron poisons or absorbers has been proposed in order to mitigate against such events. These include the use of Hf and Gd to poison the Pu and uranium as a diluent for Pu daughters. The focus of their work was to ensure compatibility of the wasteform for disposal in Yucca mountain. A similar study may be required to underpin disposal in a UK repository, however in the absence of a defined repository a number of scenarios are being modelled, the results of which will feed into work on the wasteform formulation.

Three types of criticality scenarios have been identified; in-situ, degraded and extended. The former relates to the wasteform as emplaced in the repository with the degraded mode referring to leaching of Pu from the matrix and extended to transport through the repository. Current modelling is aimed at the in-situ case and has been carried out for ceramic, glass and immobilisation MOX wasteforms. Criticality modelling for each of these wasteforms was undertaken using MONK [2] and MCNP [3] criticality modelling codes in order to verify the derived preliminary simulation k_{eff} (the neutron multiplication factor) results. The assumed geometry for the cases considered consists of 3 layers of 17 individual waste canisters placed within a larger canister and then placed into the repository. An example of the repository geometry model assumed in the simulation codes is presented in Figure 1.

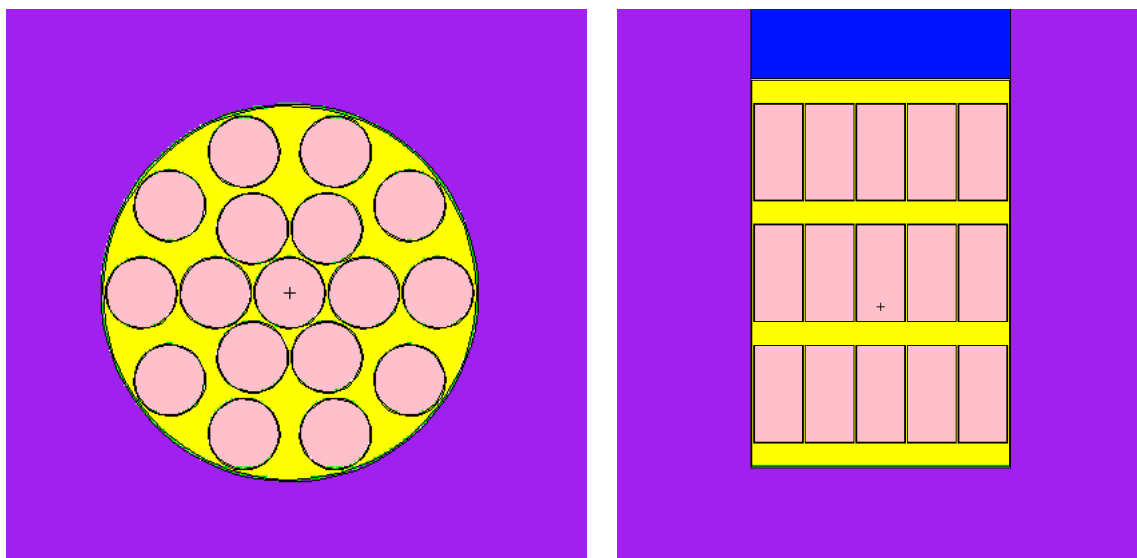


Figure 1: Assumed repository geometry model (Left: top view looking down on canisters in repository. Right: side view of in-situ canisters).

The models allow different poison strategies to be tested along with waste loading and packaging densities. Similarly the presence of interstitial materials between an array of waste canisters is being tested. Three interstitial materials under consideration are air, bentonite (a type of clay) and water. Modelling of all variants has yet to be completed but initial results show that k_{eff} can vary substantially as a result of removing neutron poisons and changing interstitial materials. The value of k_{eff} also varies depending on the source of Pu with UK Magnox derived Pu showing higher values. Currently only immobilisation MOX (12.6 w% Pu) shows k_{eff} values > 0.9 (see Figure). Note that this is for UK PWR Pu, as an example, and may well be increased should Magnox Pu be used. This programme of model simulation will be continued to evaluate the full range of Pu contents and neutron poisons for the range of possible wastefoms in more detail in future work.

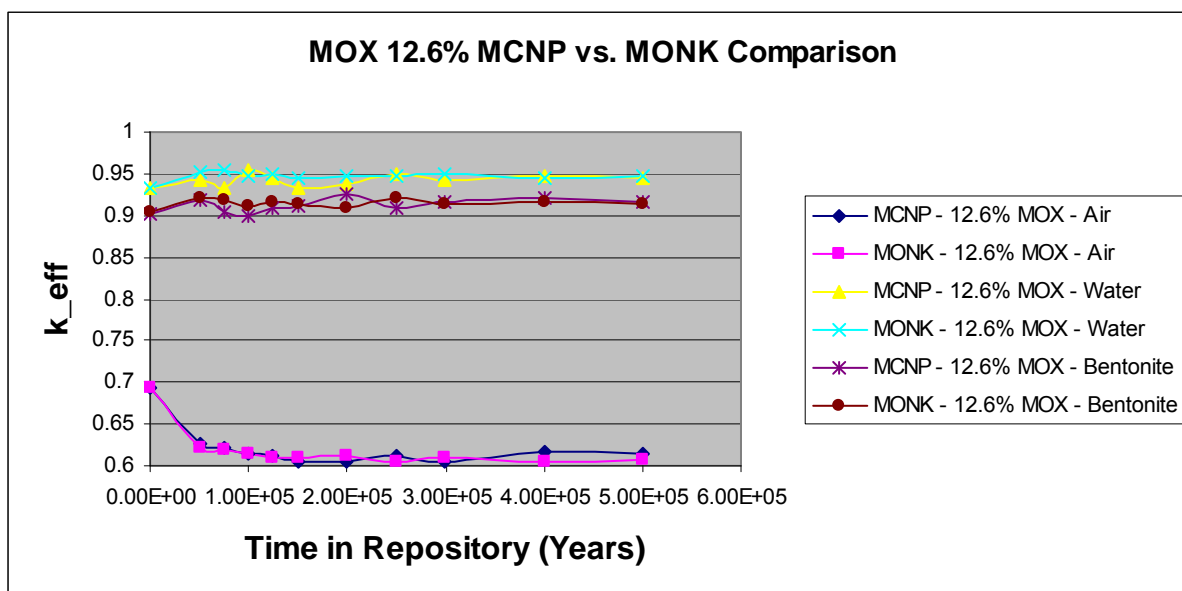


Figure 2: Graph of the preliminary modelling results for the 12.6% immobilisation MOX wasteform in repository. Comparison between the MONK and MCNP simulations is presented for three interstitial materials under consideration.

The preliminary k_{eff} results for the ceramic wastefrom currently under consideration are presented in Figure 3. Three different interstitial materials are simulated in the MONK and MCNP criticality codes to allow a verification of the results derived from the model.

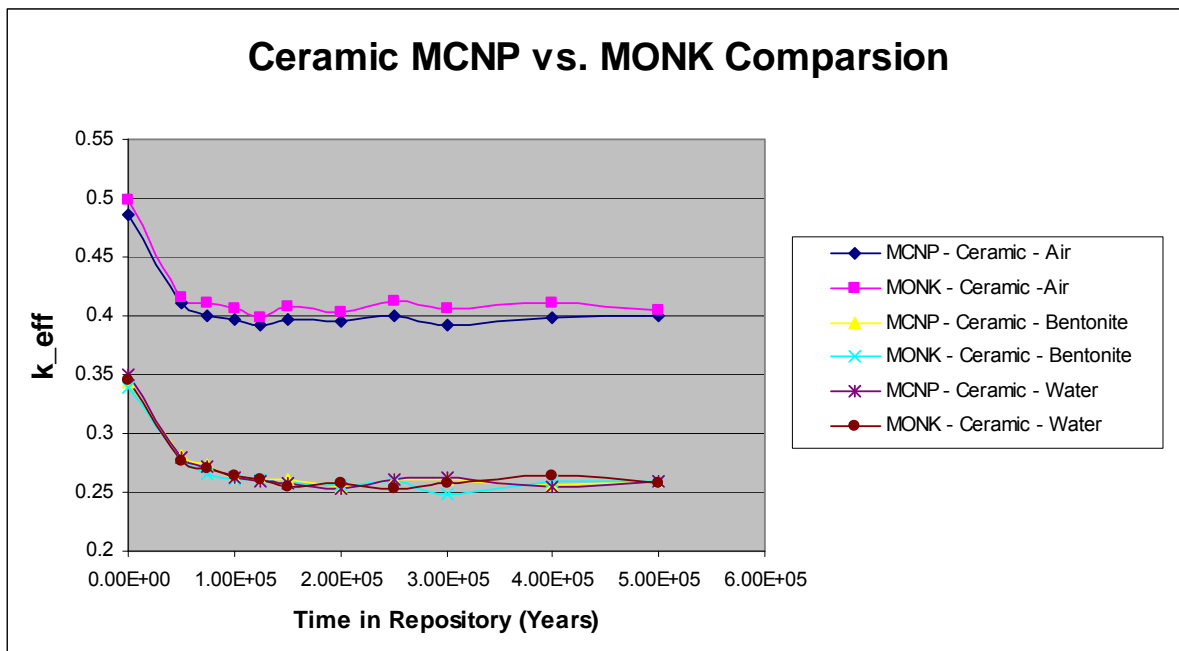


Figure 3: Graph of the preliminary modelling results for the ceramic wastefrom in repository. Comparison between the MONK and MCNP simulations is presented for three interstitial materials under consideration.

In Figure 2 the preliminary modelling results for the Alkali Tin Silicate glass composition are presented. Three different Pu incorporation rates are considered 10 w%, 15 w% and 20 w%. The interstitial material present between the canisters is assumed to be air for the k_{eff} results presented for both MONK and MCNP simulations.

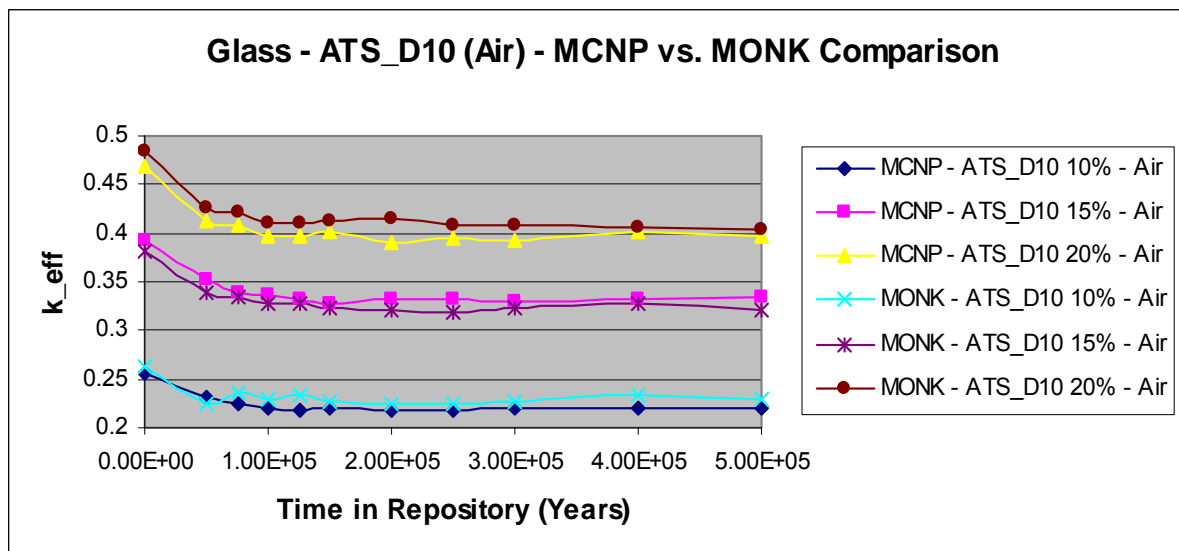


Figure 2: Graph of the preliminary modelling results for the Alkali Tin Silicate (ATS_D10) glass composition, at the three different Pu incorporation rates, with air as the interstitial material. Comparison between the MONK and MCNP simulations is presented for the materials under consideration.

4 CONCLUSIONS

This paper has summarised the progress on the Civil Pu disposition project, the benefits gained from the new knowledge and highlighted the remaining research areas that may be investigated in the subsequent stages of the project.

The National Nuclear Laboratory is conducting this project to support the NDA to allow them to advise Government on the available alternatives and the consequences of potential decisions to allow Government to make decisions on the selection of the best option(s) for civil Pu disposition in the UK. The programme so far has been aimed at underpinning credible technology options, with a strategic overview that can be used as part of the decision making process.

The work programmes investigating the various re-use strategies will provide a solid basis for evaluating the relative merits of the different UK re-use options currently under consideration.

A range of immobilisation strategies have been proposed currently covering ceramic, vitreous and MOX wasteforms. Those strategies deemed to be credible for the immobilisation of Pu will be investigated in more detail depending on decisions which will be based on several factors. Wasteform selection criteria have been proposed and will be used to highlight the most suitable options. The final decision on immobilisation options will take into account key properties of the wasteform such as aqueous durability, resistance to radiation damage, criticality mitigation and the capability to physically process both safely and cost effectively among other factors.

The modelling has been undertaken using MONK and MCNP to investigate the criticality behaviour for the various wasteforms and interstitial materials currently under consideration as part of the Pu disposition project. The preliminary results of this work will form part of the overall decision making process which will influence the selection of the best option(s) for civil Pu disposition in the UK.

REFERENCES

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