

**ICEM2009-16331**

**CONTINGENCY OPTIONS FOR THE DRYING, CONDITIONING AND  
PACKAGING OF MAGNOX SPENT FUEL IN THE UK**

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**ABSTRACT**

The UK Nuclear Decommissioning Authority (NDA) is responsible for safe and secure management of spent nuclear fuel. Magnox spent fuel is held at some Magnox reactor sites and at Sellafield where it is reprocessed using a number of facilities. It is intended that all Magnox fuel will be reprocessed, as described in the published Magnox Operating Plan (MOP) [1]. In the event, however, that a failure occurs within the reprocessing plant, the NDA has initiated a programme of activities to explore alternative contingency options for the management of wetted Magnox spent fuel.

Magnox fuel comprises metallic uranium bar clad in a magnesium alloy, both of which corrode if exposed to oxygen or water. Consequently, contingency options are required to consider how best to manage the issues associated with the reactivity of the metals. Questions of whether Magnox spent fuel needs to be dried, how it might be conditioned, how it might be packaged, and held in temporary storage until a disposal facility becomes available, all require attention. A review of potential contingency options for Magnox fuel was conducted by Galson Sciences Ltd, UKAEA and the NDA.

During storage in the presence of water, the corrosion of Magnox fuel produces hydrogen ( $H_2$ ) gas, which requires careful management. When uranium reacts with hydrogen in a reducing environment, the formation of uranium hydride ( $UH_3$ ) may occur, which under some circumstances can be pyrophoric, and might create hazards which may affect subsequent retrieval and/or repackaging (e.g. for disposal). Other factors that may affect the choice of a viable contingency option include

criticality safety, environmental impacts, security and Safeguards and economic considerations.

At post-irradiation examination (PIE) facilities in the UK, Magnox spent fuel is dried as a result of storage in air at ambient temperatures. Early French UNGG (Uranium Naturel Graphite Gaz) fuel was retrieved from pond storage at Cadarache, dried using a hot gas drying technique, oxidised and packaged in sealed canisters and placed in interim storage at the CASCAD (CASemate CADarache) facility. In the US, spent fuels including the Zircaloy clad Hanford N-Reactor fuels were cold vacuum dried and Idaho legacy aluminium clad metallic uranium fuels were hot vacuum dried; the dried fuel was then packaged in sealed and vented canisters (at Hanford and Idaho, respectively) for interim storage.

With regard to conditioning and packaging, several different approaches have been reviewed, including encapsulation in cementitious grout or polymer, high-temperature vitrification or ceramicisation, and solution in acid or alkali solution followed by cementation or vitrification (without reprocessing). All of these approaches require further research in order to be evaluated and developed further for application to formerly wetted Magnox fuel.

A variety of containers have been developed for the transport, storage and/or disposal of spent fuel in radioactive waste management programmes worldwide. Wetted Magnox spent fuel could be packaged in a container, with reservations about the potential formation of  $UH_3$  in a sealed environment where reducing conditions may develop. The applicability of different combinations of drying, conditioning and packaging

techniques to the preparation of Magnox spent fuel for long-term storage and eventual disposal are discussed.

## INTRODUCTION

Magnox fuel elements comprise a uranium metal bar clad in a sheath of Magnox (a magnesium (>99%) and aluminium alloy) [2], which is sealed and normally ‘finned’ to assist gas flow and heat transfer. The overall UK strategy that defines lifetime targets for the whole Magnox fuel cycle is the MOP (Magnox Operating Programme, currently at version 8), which covers fuel manufacture, electricity generation, fuelling and de-fuelling of reactors, and reprocessing of spent fuel [1]. According to MOP8, the existing UK Magnox spent fuel inventory (~5,500 tonnes in April 2007) will be reprocessed by 2016 [1]. It is likely for both technical and policy-related reasons that reprocessing of Magnox spent fuel will not continue in the current facilities beyond 2017. The NDA is therefore considering alternative options for Magnox spent fuel management in the event that it is not possible to reprocess the entire UK Magnox spent fuel inventory in the current facilities.

A review of potential contingency options for Magnox fuel was conducted by Galson Sciences Ltd., UKAEA and the NDA, including previous UK and overseas work on drying and subsequent dry storage of formerly wet-stored metallic uranium spent fuel (Table 1).

One possible management option would be to transfer the fuel that is currently in wet storage in ponds into interim dry storage, pending agreement on disposal options. Due to the similarities between Magnox, UNGG and Zircaloy fuels, drying methods applied at PIE facilities in the UK, the STAR (Station de Traitement, Assainissement, Reconditionnement) facility at Cadarache, and at Hanford and Idaho in the US were reviewed. Although the review mainly examined drying techniques, a range of conditioning methods and packaging types were also considered. Options for the dry storage of Magnox spent fuel are addressed in a companion paper.

**Table 1: Summary of metallic uranium fuel types reviewed and their relationship to Magnox fuel.**

Country	Programme	Fuel type	Composition of cladding	Relationship to Magnox fuel
UK	CEGB/SSEB PIE facilities Sellafield/NNL	Magnox	Magnox alloy Al 80 [3]	-
USA	Westinghouse Hanford Company (with PNNL)	Hanford N-reactor	Zircaloy	Zircaloy is less reactive than Magnox.
	Idaho National Laboratory (formerly INEEL)	Plate fuel	Aluminium	Aluminium is less reactive than Magnox.
France	CEA Cadarache	UNGG	Magnesium	Slight differences in cladding and composition.
Canada	AECL	CANDU	Zirconium alloy	There is no risk of criticality with CANDU fuel.

## ISSUES AFFECTING THE DRYING, CONDITIONING AND PACKAGING OF MAGNOX SPENT FUEL

At Magnox power stations in the UK, following discharge from a reactor, Magnox fuel is stored either in water-filled ponds or CO<sub>2</sub>-filled dry storage tubes (at Wylfa) for at least ~150 days until the heat produced by the decay of short-lived radionuclides reaches a specified level. Magnox, Zircaloy and UNGG fuels all comprise metallic uranium fuel rods surrounded by magnesium or zirconium metal alloy cladding, which is not designed for long-term wet storage. Delays to reprocessing or delays in the implementation of dry storage facilities for these fuels have resulted in extended unplanned periods of wet storage in the UK, the US and France, which causes the following problems:

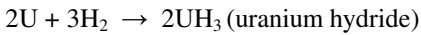
- **Cladding corrosion:** underwater, even with carefully controlled pond water chemistry, Magnox cladding is expected to corrode appreciably within five years. This leads to exposed uranium surfaces and the release of soluble fission products into the pond water.
- **Hydrogen gas generation:** uranium metal reacts with water or water vapour exothermically in the reaction:



The drying of metallic fuel under vacuum can lead to an increase in fuel temperature due to these reactions, particularly if uranium metal has been exposed by, *inter alia*, cladding degradation. There is an increased risk of thermal runaway as a result of enhanced reactivity occurring with increased operating temperatures [4]. H<sub>2</sub>

gas is potentially flammable and causes pressurisation problems if the fuel is stored in sealed containers.

- **Production of uranium hydride:** when uranium metal fuel elements with damaged or corroded cladding have been stored under sealed conditions, uranium hydride may be produced. In the absence of oxygen, the uranium first removes all the water and generates hydrogen by the reaction above. Uranium metal reacts with hydrogen in the reaction:



If subsequently re-exposed to air / oxygen, under some circumstances  $UH_3$  may spontaneously ignite.

### OPTIONS FOR DRYING OF MAGNOX SPENT FUEL

Methods have therefore been developed to dry intact and corroded wet-stored spent fuel elements from legacy programmes in a number of countries. Wet-stored fuel must be dried before it can be safely packaged for long-term dry storage or disposal. Vacuum drying and hot gas drying methods differ in that vacuum drying is initially carried out under vacuum at low gas pressures (down to 0.5 torr), whereas hot gas drying is conducted in a flow of inert gas at higher pressures.

#### Hanford Cold Vacuum Drying Process

At Hanford, an inventory of ~2100 tonnes (of uranium) of Hanford N-reactor fuel had been in storage at the K Basins, half in vented canisters (in K West basin) and half in open top canisters (in K East basin) since the early 1950s. From 1994, a project to remove the spent fuel resulted in studies on the safe drying and storage of intact and corroded fuel elements [5].

Underwater, the spent fuel was placed into baskets and loaded into a Multi-Canister Overpack (MCO), which was placed in a sealed transport cask. The first MCO was transferred to a Cold Vacuum Drying Facility (CVDF) in December 2000 [6]. The pressure inside the cask was measured on arrival. If no increase in pressure was detected, the cask headspace was vented and purged with helium to mitigate any deleterious interaction between oxygen and  $UH_3$  [7].

At the CVDF, water was removed from the MCO by draining to remove the bulk water, then cold vacuum drying for 24 to 48 hours to remove free water [5]. The dried MCOs were then shipped to a Canister Storage Building for secure interim storage.

#### Idaho Hot Vacuum Drying and Passivation Process

Idaho National Engineering and Environmental Laboratory (INEEL, now INL) used a combination of vacuum drying and heating to remove free water from spent fuel that had been wet-stored in a storage basin (Chemical Processing Plant 603 Basin). Eight fuel types (including sodium bearing, uranium metal and aluminium clad spent fuel) were dry-canned in the 1990s [8]. The spent fuel was loaded underwater into self-draining fuel buckets, which were remotely loaded into a new

stainless steel canister (0.45 m external diameter x 3.4 m long) and transferred into the Fuel Canning Station.

The canister was placed within a rotating heated insert. A temporary vacuum lid, a pressure transducer and two thermocouples were placed onto the canister flange O-ring and the vacuum drying process was started [8,9]. The canister was heated to a constant internal temperature of 50°C and the vacuum pump was set to ensure that the canister pressure remained above 5 torr (the triple point of water at 0.01°C); if the pressure dropped below this point the water froze, slowing the drying process and potentially damaging the fuel [8]. When the internal canister pressure dropped to match the pressure at the inlet to the vacuum pump, the system was isolated and the pressure/temperature ratio monitored for 1 to 2 hours. If the pressure did not remain constant, the drying process would be repeated [8].

Once free water was removed, the temperature of the fuel inside the canister was increased to 150 °C for two hours, with an inert gas system providing a low pressure argon (with 2% oxygen) flow to passivate uranium hydride and reduce the likelihood of pyrophoric fuel [8]. Increasing the temperature to 150°C removed all free water and much of the physi-sorbed water in the corrosion products and clay sediments. The canisters were fitted with vented lids [8], which prevented the formation of uranium hydride, and also prevented over-pressurisation of the storage container by venting any hydrogen gas produced [9].

#### CEA Cadarache Hot Gas Drying Process

In France, CEA planned to remove and reprocess a wide range of spent fuels in long term storage. Twenty tonnes of Uranium Naturel Graphite Gaz (UNGG) spent fuel from early French gas-cooled reactors, similar in design to the UK Magnox stations, was stored for over 25 years in steel canisters in two water-filled ponds at the INB 56 facility in Cadarache [10]. A significant proportion of these canisters contained uranium hydride corrosion products, which spontaneously ignited on contact with air. The fuel therefore needed treatment to ensure safe transport prior to either reprocessing at Marcoule (until 1997) or storage at CASCAD (dry vault storage facility at Cadarache) [11]. The canisters were transferred to an underwater monitoring station to assess the state of the fuel and detect water ingress into the canister. When the canister was removed from the pond, a transfer hood containing argon was used to prevent contact with air while the canister was wrapped and packaged in shielded casks for transport to the STAR (Station de Traitement, Assainissement, Reconditionnement) facility [10].

The STAR facility contained three stainless steel-lined inspection cells with 1.2m thick walls, which provided containment and shielding [12]. The magnesium cladding was mechanically separated from the uranium metal. De-cladding (also termed de-canning) removes carbonaceous deposits, prevents pressurisation issues, increases packing densities by a factor of five, and promotes the removal of water by exposing

uranium metal [13]. To dry and condition the uranium metal, it was placed in a furnace and heated to 100°C in argon to remove liquid water and prevent further uranium – water interactions. The uranium was then heated to 430°C in argon gas at atmospheric pressure to pyrolyse uranium hydride ( $\text{UH}_3 \rightarrow \text{U} + \frac{3}{2} \text{H}_2$ ) [10,12]. As the metallic uranium particles are also flammable, they were partially oxidised at 250°C, under varying pressure with the proportion of  $\text{O}_2$  gradually increased from 5% to 21% (the amount found naturally in air) until a stable product was obtained [11,12].

The stabilised uranium was placed in aluminium alloy (AG3) canisters with soluble magnesium windows, designed to meet both road transport and Marcoule UP1 reprocessing plant requirements, and welded [10]. The sealed canisters were transferred to a second less contaminated cell in a nitrogen atmosphere for weighing, decontamination if required and leaktightness checks. The canisters were transferred to a third clean cell for storage prior to transportation in shielded casks (each holding 8 to 12 canisters in baskets) [10].

### **Post-Irradiation Examination (PIE) Facility experience**

At PIE facilities in the UK, wet-stored irradiated Magnox fuel is passively dried as a result of long-term storage in air at ambient temperatures [14]. Air-dried corroded Magnox spent fuel has been subsequently stored in air at PIE facilities for extended periods.

### **OPTIONS FOR CONDITIONING OF MAGNOX SPENT FUEL**

Under some conditions, Magnox spent fuel requires active management because both magnesium and uranium metal react in air and water. It is therefore uncertain whether it would be accepted for direct disposal in a geological disposal facility without conversion to a stable chemical form [15]. If, in a contingency situation, an interim storage strategy is chosen to manage the UK Magnox spent fuel inventory, consideration should also be given to the ultimate fate of the spent fuel and particular care taken not to foreclose options.

Options for the treatment of Magnox spent fuel may include:

- Inorganic encapsulation with cements or geopolymers
- Organic polymer-based encapsulation
- Chemical treatment, followed by cementation or vitrification
- Alternative methods, including electrometallurgical treatment and transmutation

Inorganic and organic polymer-based encapsulation methods have previously been used to encapsulate Magnox swarf and Magnox Fuel Element Debris (FED) on an industrial scale. Chemical treatment of spent fuel, which includes reprocessing, has been conducted on an industrial scale at the Magnox Reprocessing Plant in Sellafield. Trials of

electrometallurgical treatment methods were conducted on Hanford N-reactor fuels.

### **Inorganic Encapsulation Methods**

Magnox swarf from de-canning operations at Sellafield, which is predominantly Magnox cladding with uranium carry-over of between 1 to 3% of the original fuel mass [16], has been routinely packaged in the Magnox Encapsulation Plant (MEP) at Sellafield since 1990 [2]. In total, more than 23,000 cementitious packages containing Magnox and aluminium Intermediate Level Waste (ILW) will be produced by the MEP [17]. Magnox swarf was initially stored underwater, then transferred to a 0.5m<sup>3</sup> drum and dewatered in an inert atmosphere. A blend of blast-furnace slag (BFS) and ordinary Portland cement (OPC) (in a 3:1 ratio) grout, having a lower free water content, was pumped into the drum and vibrated to ensure complete infilling of the swarf, then set within 24 hours and cured. The drum was capped with cement, cured and sealed [2]. Rate-limited reactions have been observed between Magnox swarf and the OPC-based cementitious grout [17]. Corrosion of cladding materials, including Al, Mg and Magnox alloys, in a high pH cement environment (~12.5 for OPC) generates hydrogen gas and produced layers of corrosion products that occupy a larger volume than the original waste, which with extended storage may cause some cracking of the waste form [17].

Further trials on intact and damaged Magnox spent fuel or uranium metal would be required before this method could be considered for the large-scale encapsulation of spent fuel.

### **Polymer-based Encapsulation Methods**

Thermosetting polymers, such as epoxy or polyester resin, have not been used to encapsulate spent fuel on an industrial scale, and their long-term performance is not as well established as that of OPC-based grout; however, this technology has been extensively tested in the US for encapsulation of resins [18].

Polymer-based encapsulation has also been examined as an alternative to cement for Magnox cladding waste (generated from decanning activities, or from desplitting (removal of fins etc. to improve packing density for transport)) at Trawsfynydd Nuclear Power Station. Polymer is being considered as an alternative to cement because of its superior flow properties for infilling small void spaces [19] and Magnox metal encapsulated in VERI polymer passed fire tests [20]. Disadvantages of thermosetting plastics are that they set relatively quickly, which may be problematic when operating on an industrial scale. Some polymers are expensive and their use would increase consumption of oil feedstock, which is a limited resource.

### **Chemical Treatment**

Magnox spent fuel is currently reprocessed at Sellafield using the PUREX (Plutonium URanium EXtraction) process. The full PUREX process starts with acid dissolution and results in the separation of U, Pu and long-lived fission products [21]. Chemical treatment of Magnox spent fuel, which could be

performed with or without the Magnox cladding, by dissolution in acid or enhanced corrosion under alkaline conditions was proposed by CEGB/SSEB [22] to convert spent Magnox fuel into a chemically stable form. Enhanced corrosion could be achieved by maintaining alkaline conditions at an elevated temperature, producing sludge. The resulting products could be cemented or vitrified to produce a waste form that would gain acceptance for disposal in a geological repository. Such processes are effective at producing a homogeneous waste form that can be easily assayed.

Spent fuel reprocessing using the PUREX process is a mature technology; at the end of 2003, more than 89,000 tHM of commercial spent fuel had been reprocessed, largely at the two commercial plants at La Hague and Sellafield [23].

Vitrified waste products are internationally recognized and licensed as an acceptable waste form [23]. Highly Active Liquid Waste (HALW) from nuclear fuel reprocessing has been treated at the Waste Vitrification Plant (WVP) at Sellafield since 1991. The Magnox fuel would be dissolved and the solution concentrated by evaporation, but not neutralised. The liquor would then be transferred to a calciner with glass making components and heated to 1100°C. The molten glass would be poured into a refractory lined 500 litre drum and allowed to cool. The inclusion of uranium and plutonium in the vitrified product, which are currently separated using the standard PUREX process, would require development work and greater precautions would have to be taken to prevent criticality during waste processing [22].

### **Electrometallurgical Treatment**

Electrometallurgical separation in an alkali metal chloride liquid was proposed as a method to separate uranium and transuranic fission products from uranium metal Hanford N-reactor fuels. This method could be used without any pre-treatment for undamaged fuel elements; however, failed fuel elements would have to be treated by head-end processing including fuel disassembly, chemical purification and oxide reduction. Following separation, the fission products and transuranic elements would be immobilised by blending waste salt with zeolite and borosilicate glass at high temperature (850 – 900°C) and pressure (14,500 – 25,000 psi) to produce a vitrified waste form. The uranium would be melted at 1600°C and cast into ingots, in an argon atmosphere [24].

### **OPTIONS FOR PACKAGING MAGNOX SPENT FUEL**

Magnox spent fuel may be packaged in sealed or vented containers or, following drying, stored bare in a suitably designed vault store. In addition to the sealed Hanford MCO, vented INEEL canister and French aluminium alloy (AG3) canisters discussed above, a number of containers are commercially available:

- CEA Spent Fuel Holder
- SKB Spent Fuel Canister
- NAGRA Spent Fuel Canister

This is not a comprehensive list, but is intended to highlight distinct types of containers that may be suitable for the interim storage, and potentially the eventual disposal, of Magnox spent fuel.

## **DISCUSSION**

### **Drying Options**

Drying of wetted Magnox spent fuel, particularly if fuel has been stored in water for extended periods, is complicated by the reactivity of Magnox and uranium metal and the potential for generation of pyrophoric corrosion products and hydrogen gas. Post-irradiation examination experience in the UK demonstrates that air-dried corroded Magnox spent fuel can be subsequently stored in air for extended periods without significant degradation.

UNGG and Magnox fuels both have magnesium cladding; however, Zircaloy and aluminium are less reactive under the conditions commonly encountered in the nuclear industry. Because the cladding materials and uranium metal are chemically reactive in water and air, sufficient water must be removed to prevent hazardous conditions developing from the production of hydrogen gas and potentially pyrophoric corrosion products. The definition of 'sufficiently dry' depends upon whether the fuel is being prepared for passive storage or further treatment. Fuel being dried prior to storage in a sealed container will have a bounding limit for pressure build-up within the container (e.g. Hanford MCO) by the generation of hydrogen gas and expansive corrosion products. Storing fuel in vented containers (e.g. INEEL canisters) will prevent the formation of uranium hydride, and also prevent over-pressurisation of the storage container by venting hydrogen gas, should it be produced. At Hanford, it was assumed that the sealed MCOs would be disposed of in a geological disposal facility and not reopened. This reduced the significance of uranium hydride production, as no oxygen would enter the MCO in the future. In the UK, sealed containers of unpassivated spent fuel may not be accepted for disposal.

The Hanford cold vacuum drying method had similar time and throughput constraints (four years from start of operations) to those anticipated if any Magnox contingency plans needed to be implemented [6].

The Cadarache hot gas drying process decomposed uranium hydride and oxidised metallic uranium surfaces, reducing the reactivity of the fuel in interim dry storage [10]. This minimises future hydrogen gas and uranium hydride production and therefore both reduces the risk of over-pressurisation of the sealed canister and mitigated the future hazards associated with reopening the canisters if inspection or repackaging are required. However, the Cadarache method was energy-intensive and time-consuming and would require a number of parallel facilities to treat the UK Magnox spent fuel inventory, which may not be accommodated at the Sellafield site.

All of the drying methods reviewed could potentially be used to treat both intact and corroded or broken Magnox fuel elements. However, none of the methods has established that the fuel itself reaches the temperatures required to produce a sufficiently dry product. Decay heat or direct heating of the fuel elements (for example, by immersion in hot water) would improve evaporative drying at <100°C or facilitate the fuel reaching temperatures >100°C for positive drying [13].

### **Conditioning and Packaging Options**

The review of spent fuel conditioning and packaging options revealed that there is little operational experience of encapsulation of formerly wet-stored spent fuel.

Of the various approaches that have been considered for conditioning Magnox and other spent fuels, all have potential difficulties. Encapsulation of spent fuel in cementitious grout has been studied in various trial experiments at Sellafield and results suggest that it cannot presently be considered as a passively safe, stable waste form. Encapsulation in organic polymer has been less well studied but may be a more favourable encapsulant, as polymers have lower water/solids ratios than OPC-based grouts. Dissolution or enhanced corrosion destroys chemical reactivity and potentially allows the resulting solution to be converted to a stable waste form by vitrification or cementation. However, the process would be complex and require extensive research and development to implement.

A variety of containers have been developed for the transport, storage and/or disposal of spent fuel in radioactive waste management programmes worldwide and could be considered in association with any of the drying and conditioning options described in this report.

### **SUMMARY AND FURTHER WORK**

Drying of wetted Magnox spent fuel, particularly if fuel has been stored in water for extended periods, is complicated by the reactivity of Magnox and uranium metal and the potential for generation of pyrophoric corrosion products and hydrogen gas. At PIE facilities in the UK, Magnox spent fuel is routinely passively dried in air at ambient temperatures. Early French UNGG fuel was retrieved from pond storage at Cadarache, dried using a hot gas drying technique, oxidised and packaged in sealed canisters and placed in interim storage at the CASCAD facility. In the US, spent fuels including the Zircaloy clad Hanford N-Reactor fuels and Idaho legacy aluminium clad metallic uranium fuels were dried by cold and hot vacuum drying respectively and packaged in sealed canisters for interim storage.

With regard to conditioning and packaging, several different approaches have been reviewed, including encapsulation in cementitious grout or polymer and solution in acid or alkali solution followed by cementation or vitrification (without reprocessing). All of these approaches require further research in order to be evaluated and developed further for application to formerly wetted Magnox fuel.

### **ACKNOWLEDGMENTS**

The Nuclear Decommissioning Authority funded this work through its Direct Research Portfolio under contract CJ000102.

We are grateful to the participants of an expert workshop, held by GSL and the NDA on 22<sup>nd</sup> January 2009 in Warrington, who reviewed the international literature review and provided context and additional information.

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