MANAGEMENT OF UKAEA GRAPHITE LIABILITIES

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Abstract. The UK Atomic Energy Authority (UKAEA) is responsible for managing its liabilities for redundant research reactors and other active facilities concerned with the development of the UK nuclear technology programme since 1947. These liabilities include irradiated graphite from a variety of different sources including low irradiation temperature reactor graphite (the Windscale Piles 1 & 2, British Energy Pile O and Graphite Low Energy Experimental Pile at Harwell and the Material Testing Reactors at Harwell and Dounreay), advanced gas-cooled reactor graphite (from the Windscale Advanced Gas-cooled Reactor) and graphite from fast reactor systems (neutron shield graphite from the Dounreay Prototype Fast Reactor and Dounreay Fast Reactor).

The decommissioning and dismantling of these facilities will give rise to over 6,000 tonnes of graphite requiring disposal. The first graphite will be retrieved from the dismantling of Windscale Pile 1 and the Windscale Advanced Gas-cooled Reactor during the next five years.

UKAEA has undertaken extensive studies to consider the best practicable options for disposing of these graphite liabilities in a manner that is safe whilst minimising the associated costs and technical risks. These options include (but are not limited to), disposal as Low Level Waste, incineration, or encapsulation and disposal as Intermediate Level Waste.

There are a number of technical issues associated with each of these proposed disposal options; these include Wigner energy, radionuclide inventory determination, encapsulation of graphite dust, galvanic coupling interactions enhancing the corrosion of mild steel and public acceptability.

UKAEA is currently developing packaging concepts and designing packaging plants for processing these graphite wastes in consultation with other holders of graphite wastes throughout Europe.

"Letters of Comfort" have been sought from both the Low Level Waste and the Intermediate Level Waste disposal organisations to support the development of these waste disposal strategies. UKAEA is liasing closely with the disposal organisations to ensure that all their concerns are addressed satisfactorily.

This paper will describe:

i. the varied nature, inventory and history of UKAEA's graphite liabilities;
ii. the options that have been considered for the long-term storage and disposal of UKAEA's graphite liabilities;
iii. the technical issues that have been considered in the development of these options;
iv. the recent developments in the consideration of Wigner energy.

The paper will summarise the current status and future plans of the UKAEA graphite waste packaging strategies, and describe the interactions with the disposal organisations.

1. INTRODUCTION

The UK Atomic Energy Authority (UKAEA) has built and operated a wide range of nuclear facilities since the late 1940's for the development of all aspects of atomic energy including reactor systems, fuel and reprocessing technology. These facilities, located on our sites at Culham, Dounreay, Harwell, Windscale and Winfrith are varied and complex. UKAEA has been tasked with managing the liabilities from these nuclear facilities.

These liabilities include graphite from a number of redundant reactors. Stage 3 decommissioning and dismantling of these facilities will give rise to over 6,000 tonnes of graphite requiring disposal. UKAEA has undertaken extensive studies to consider the best practicable options for disposing of these graphite liabilities in a manner that is safe whilst minimising the associated costs and technical risks.

This paper describes the varied nature, inventory and history of UKAEA's graphite liabilities and the options that have been considered for the long-term storage and disposal of these liabilities. The paper outlines the technical issues that have been considered in the development of these options and the recent developments in making the technical case for the packaging and disposal of graphite containing low irradiation temperature Wigner energy.

2. BACKGROUND

This section provides details on the design, construction and operating history of those reactors containing graphite liabilities. It also provides details on the current status of decommissioning these reactors, and outlines the packaging and disposal strategies and the associated technical issues. The basis of these strategies and detail of the technical issues are discussed in later sections. The data presented in this section is summarised in Tables I and II.

TABLE I. SUMMARY OF REACTOR HISTORY

Reactor	**Type of Reactor**	**Operations Began**	**Operations Ceased**	**Power Output (Th.)**	**Status of Decommission-ing**
GLEEP	Research reactor	1947	1990	3-60 kW	Awaiting Stage 3 decomm.
BEPO	Research reactor	1948	1968	6 MW	Awaiting Stage 3 decomm.
Windscale Pile 1	Plutonium production	1950	1957	180 MW	Awaiting Stage 2/3 decomm.
Windscale Pile 2	Plutonium production	1950	1957	180 MW	Awaiting Stage 2/3 decomm.
DFR	Research reactor – fast reactor systems	1959	1977	60 MW	Currently in Stage 1 decomm.
PFR	Research reactor-commercial fast reactors	1974	1994	600 MW	Currently in Stage 1 decomm.
DIDO	Research reactor – material testing reactor	1956	1990	10-26 MW	Awaiting Stage 3 decomm.
PLUTO	Research reactor – material testing reactor	1957	1990	10-26 MW	Awaiting Stage 3 decomm.
DMTR	Research reactor – material testing reactor	1958	1969	10-26 MW	Awaiting Stage 2/3 decomm.
WAGR	Research reactor – commercial AGRs	1962	1981	105 MW	Currently in Stage 3 decomm.

2.1. Graphite Low Energy Experimental Pile

The Graphite Low Energy Experimental Pile (GLEEP) was built at Harwell in the mid-forties for research purposes, and operated between August 1947 and September 1990. GLEEP was a thermal heterogeneous reactor; graphite moderated and reflected, and air-cooled. The reactor is formed from interlocking graphite blocks to give a graphite cube of 6.4 m side, with 682 fuel channels in three radial zones. The core used two types of fuel; aluminium sprayed uranium metal and pelleted UO_2 in cans, however for most of its operating life the fuel was a natural uranium metal bar encased in an aluminium can. The GLEEP core contains 505 tonnes of graphite and is shielded by 1.5 m of barytes concrete [1].

TABLE II. SUMMARY OF GRAPHITE DATA

Reactor	**Quantity of Graphite (te)**	**Purpose of the Graphite**	**Major Radionuclides**	**Temperature of Irradiation (^{o}C)**
GLEEP	505	Moderator and reflector	H-3, C-14 & Eu-152	18-15
BEPO	766	Moderator and reflector	H-3 & C-14	40
Windscale Pile 1	1966	Moderator and reflector	H-3, C-14 & Cs-137	20-150
Windscale Pile 2	1966	Moderator and reflector	H-3 & C-14	20-150
DFR	200	Borated graphite neutron shield	To be assessed	350
PFR	To be assessed	Neutron shield	To be assessed	560-600
DIDO	17	Reflector only	H-3 & C-14	108
PLUTO	17	Reflector only	H-3 & C-14	108
DMTR	17	Reflector only	H-3 & C-14	108
WAGR	210	Moderator and reflector	H-3 & C-14	300-350

Initially, GLEEP was intended for use as a precision test reactor for basic neutron physics measurements and for routine quality testing. However for the first 18 months of operation it was used as a radioisotope producer until the British Energy Pile O (BEPO) reactor was completed. GLEEP operated at an average temperature of 18 ^{o}C and a minimum temperature near the charge face of the reactor of 15^{o}C. After its initial start-up GLEEP operated for over a year at an average power of 60 kW (Th.) until January 1949, when the fuel was replaced, and then at 3 kW (Th.) until its closure. The normal operating cycle for GLEEP was 12 days at power and two days for shutdown for maintenance, thus the irradiation history is equivalent to 1.644E5 kWh (Th.) and as the thermal flux in the centre of the GLEEP core was 1E9 n.cm^{-2} s^{-1} at 3kW, the total neutron dose was 1.97E18 n.cm^{-2} [3].

The calculated radionuclide inventory of the GLEEP graphite is dominated by H-3 at 95.1 GBq, C-14 at 7.87 GBq and Eu-152 at 5.95 GBq of graphite. A core sample of the graphite will be taken in the next 12 months to confirm the radionuclide inventory.

Following closure and defuelling of the reactor GLEEP has been held under care and maintenance at the Harwell site awaiting stage 3 decommissioning which is planned to be undertaken in the next five years [2]. A number of option studies have been undertaken to establish the optimum solution for the disposal of the GLEEP graphite from the core. These studies have considered incineration, burial, disposal as ILW and disposal as LLW. The current strategy, whilst awaiting the results of the core sample, is that the core will be dismantled and loaded into half-height ISO containers for disposal at the UK Low Level Waste disposal facility, Drigg in Cumbria. Under this option the main technical issues to be addressed are the demonstration of the low levels of Wigner energy and the carbon-14 content of the waste.

2.2. British Experimental Pile O

Following the construction of GLEEP a second heterogeneous thermal reactor was built at Harwell in the 1940s, and operated from July 1948 until December 1968. Known as British Experimental Pile O (BEPO) it was the first large, fully engineered reactor in Europe. Again BEPO was uranium fuelled, graphite moderated and reflected, and air-cooled. The reactor was also made from interlocking graphite blocks to form a graphite cube of 7.9 metre side, with 888 horizontal fuel channels on a 184 mm square pitch. The fuel was aluminium clad natural or slightly enriched uranium. The BEPO core contains 766 tonnes of graphite [1].

BEPO provided valuable reactor experience leading to the successful development of Britain's first and second nuclear power programmes. It also provided a regular supply of radioisotopes for use in medicine and industry. BEPO operated at a power output of 6 MW (Th.) for most of its 20 year service. During its operational life stored Wigner energy in the core graphite was annealed out in 1954, 1958 and again in April 1961. In 1961 the air inlet temperature was raised from 40°C to 45°C to provide some self-annealing of the stored energy.

Tritium and C-14 dominate the radionuclide inventory of the BEPO graphite. The high levels of carbon-14 and its long half-life mean that the activity will remain high enough so that for any decommissioning plan in the foreseeable future the graphite in the core will need to be treated as ILW.

Following closure and defuelling of the reactor BEPO has been held under a care and maintenance regime awaiting Stage 3 decommissioning [2]. A detailed option study is currently being undertaken to consider the options for decommissioning. This study is considering continued decay storage of the steel associated with the core, incineration and repackaging of the graphite into Nirex boxes for disposal. BEPO is virtually unique in that a 6 inch core sample has been taken from the reactor face to its centre. This core is currently stored on the Harwell site and will enable profiling of the Wigner energy and radioactive inventory to be undertaken.

There a number of technical issues associated with the packaging for disposal of BEPO graphite. The main areas of concern are the levels of Wigner energy and the activity associated with the graphite.

2.3. Windscale Piles

The two Windscale Piles (Pile 1 and Pile 2) were air-cooled, graphite moderated and reflected reactors built in the late 1940s for the production of plutonium. Each Pile consisted of a graphite core, roughly in the shape of a horizontal cylinder 15.32 m diameter by 7.43 m deep. Each Pile was built from blocks of graphite keyed together by graphite slats. The slats also displaced the graphite blocks to produce gaps between blocks in the two horizontal planes. The moderator contains 3444 fuel channels and 909 isotope channels, all of which are horizontal. There are 1966 tonnes of graphite in the Pile 1 core and the same in Pile 2. The power output of each of the Windscale Piles was 180 MW (Th.) [1].

The operators were aware of the stored Wigner energy hazard in the graphite and so controlled releases were carried out at regular intervals to relieve the graphite moderator of the stored energy that had accumulated as a result of neutron irradiation. To release the Wigner

energy the air blowers were shut down and the core heated up by a controlled criticality excursion. This heating initiated the Wigner release that caused the graphite temperature to increase further, thereby indicating that the release had occurred, and the fans were restarted and the control rods inserted to reduce the reactivity to normal levels.

The Windscale Piles operated from October 1950 to October 1957 at which time a fire occurred in Pile 1 during a routine Wigner energy release. Following the successful extinguishing of the fire, both Pile 1 and Pile 2 were shut down and the fuel and isotope cartridges that could be readily removed, were cleared from the Piles. Some fuel and isotope cartridges remain within the fire-affected zone in Pile 1; Pile 2 was cleared of fuel, but some isotope cartridges remain in the core.

The activity in the graphite is dominated by C-14, Cs-137 and tritium. The graphite has an average specific activity of 57 GBq/te (outside the fire-affected zone in Pile 1).

Since October 1957 the Piles have been maintained on a care and maintenance basis. Stage 1 decommissioning of both reactors has been completed and Stage2/3 decommissioning of Pile 1 will commence shortly. Stage 2/3 decommissioning of Pile 2 has been deferred because there is no safety driver to undertake the decommissioning of Pile 2 at the current time [2].

To aid the disposal strategy the graphite from Pile 1 has been divided into two categories: graphite outside the fire-affected zone defined as undamaged graphite; and graphite from the fire-affected zone defined as damaged graphite. 'Damaged' and 'undamaged' do not refer to the physical integrity of the graphite but to the location of that graphite in the core. All of the graphite is classified as ILW. It is proposed to package the undamaged graphite in the Nirex 4 m ILW boxes and the damaged graphite into 3 m^3 boxes.

There are a number of technical issues associated with the packaging of graphite for disposal from Windscale Piles 1 and 2. The main areas of concern are Wigner energy in the graphite, the encapsulation/solidification of dust from both dismantling of the graphite and from the fire affected zone, and the activity associated with the graphite.

2.4. Dounreay Fast Reactor

The Dounreay Fast Reactor (DFR) was built at Dounreay starting in 1955 to establish the feasibility of the fast breeder system and to provide information and operating experience needed to design a prototype reactor for full scale power production. DFR operated between 1959 and 1977. The reactor core is hexagonal, 0.53 m high and 0.52 m wide, containing 324 fuel channels. The core was cooled using a liquid sodium/potassium alloy with an outlet temperature of 350^{o}C. The fuel consists of an annular cylinder of 75% enriched uranium alloy, clad internally and externally with niobium. Originally U/Cr alloy was used as the fuel which was later replaced to U/Mo [1].

Following the first criticality in 1959 the reactor was operated at very low power levels for several months and then progressively increased over the following three years until it reached the design power of 60 MW (Th.) in July 1963.

The outside of the reactor tank is surrounded by a 1.2 m thick borated graphite jacket, which provides a thermal neutron reflector and shield. This jacket contains approximately 200 tonnes of borated carbon.

DFR is currently undergoing stage 1 decommissioning. Removal and destruction of the primary and secondary sodium/potassium coolant will be undertaken in the near future. As part of Stage 3 decommissioning the graphite from the neutron shield will be cut into suitable lengths and packaged into standard Nirex 4 m ILW boxes for disposal to the deep waste repository. The issues associated with the disposal of the graphite from DFR are residual sodium/potassium associated with the graphite and the presence of any unusual radionuclides of significance to Nirex.

2.5. Prototype Fast Reactor

Based on the design and operating experience gained from DFR the Prototype Fast Reactor (PFR) was built at Dounreay in the 1960/70s to obtain information to support the design of commercial fast reactors. It operated between March 1974 and March 1994. The PFR consists of a single stainless steel tank 12 m in diameter by 15 m deep, containing the entire primary circuit, breeder blanket and graphite shielding, 6 intermediate heat exchangers and three pumps for circulating the primary sodium coolant. The fuel consists of mixed PuO_2/UO_2; the inner core contained 19% Pu and the outer core 25% Pu. The design power of PFR was 600 MW (Th.) [1].

The neutron shielding comprises of seven rows of neutron shield rods, consisting of mild steel tubes filled with mild steel or graphite cores of varying diameters and lengths.

PFR is currently undergoing Stage 1 decommissioning with the recent removal of the control rods from the core. Removal and destruction of the primary and secondary sodium coolant is due to commence shortly. During Stage 3 decommissioning the neutron shield rods will be cut into standard lengths and packaged into Nirex 4 m ILW boxes for disposal to the deep waste repository. The issues associated with the disposal of the graphite from PFR are the possible presence of residual sodium associated with the graphite, the activity associated with the mild steel and the presence of any unusual radionuclides of significance to Nirex.

2.6. Material Testing Reactors

UKAEA is currently managing the liabilities of three redundant materials testing reactors of essentially the same design; namely DIDO and PLUTO both at Harwell, and DMTR at Dounreay. All of these reactors were built and started operation in the late 1950's DMTR was shut down in 1969, DIDO and PLUTO kept operating until 1990. The MTRs used a highly enriched uranium/aluminium alloy fuel. They were heavy water moderated and cooled thermal reactors with a graphite reflector. The reactors operated at an initial power output of 10 MW (Th.), which was increased in stages to 26 MW (Th.) (for DIDO and PLUTO) after later fuel modifications [1].

There are 17 tonnes of graphite associated with the reflectors in each of the reactors. The total activity associated with the reflector from each of the MTRs is 17 TBq (total) which is dominated by the presence of C-14 and tritium.

All the MTRs were defuelled immediately after operations ceased. Stage 1 and 2 decommissioning has now been completed on DIDO and PLUTO and the reactors are currently being held under a care and maintenance regime to allow for decay of cobalt-60 before stage 3 decommissioning will be undertaken. Stage 1 decommissioning of DMTR has been completed and this reactor is currently being held under care and maintenance awaiting Stage2/3 decommissioning in the future. The plan is that a dedicated packaging and treatment plant will be constructed inside the reactor dome. The graphite associated with the reflector will be packaged into Nirex 4 m ILW boxes.

Again the technical issues associated with the packaging of this graphite are Wigner energy, dust and flotation of the graphite during grouting.

2.7. Windscale Advanced Gas Cooled Reactor

The Windscale Advanced Gas-cooled Reactor (WAGR) was built to study the advanced gas-cooled power reactor system; to prove in service performance of fuel suitable for a commercial reactor; to serve as a test bed for further development of fuel and other components; and to provide operational experience of power production [1].

Building of the WAGR started in the late 1950s, and it operated between 1962 and 1981. WAGR was a carbon dioxide cooled, graphite moderated and reflected reactor using uranium dioxide fuel in stainless steel cans. The reactor consists of a graphite moderator 4.5 m in diameter and 4.26 m high, housed in a cylindrical reactor vessel with hemispherical ends. The core contains 210 tonnes of graphite in approximately 3000 bricks. There are 200 fuel channels, each of which contained a standard fuel stringer consisting of four elements, each of 2 clusters of 21 pins. The reactor was designed to operate isothermally at a temperature of 350 ^{o}C, with a power output of 105 MW (Th.)

Stage 1 decommissioning involving fuel removal operations commenced immediately after shutdown in 1981. Further decommissioning operations and plant modifications which have been carried out to date have involved the removal of structures, equipment and services external to the reactor and these activities essentially form part of Stage 2 decommissioning. Initial Stage 3 operations have been carried out with the removal of the reactor top dome and associated components, and full-blown stage 3 decommissioning will commence shortly.

The graphite waste from the Stage 3 decommissioning will be packaged in concrete WAGR boxes for disposal as ILW. Because of the high operating temperature Wigner energy is not considered to be of concern for the packaging or disposal of the moderator and reflector graphite. However there are a number of other areas of concern for the packaging of this waste that are common to all the graphite wastes including the encapsulation of graphite dust in cement, and problems of flotation of graphite in the encapsulation grout.

3. OPTIONS FOR DISPOSAL OF UKAEA GRAPHITE LIABILITIES

UKAEA has undertaken extensive option studies and technical assessments to consider the best practicable options for disposing of these graphite liabilities in a manner that is safe whilst minimising the associated costs and technical risks. These options have been evaluated

via a number of different methods including option studies, Kepner Tregoe analysis and value engineering studies, against such criteria as:

- public acceptability;
- minimisation of environmental discharges;
- ease of regulator acceptance;
- makes best use of available technology;
- minimises production of secondary wastes;
- minimises overall dose uptake;
- minimises overall lifetime costs.

This section of the paper considers some of these options in more detail.

3.1. Low Level Waste

Graphite from a number of the reactors is classified as LLW on the basis that their gross activity is below the 4GBq/te alpha and 12 GBq/te beta/gamma limit. However, there are additional radiological constraints placed on the disposal of LLW by the UK LLW disposal organisation BNFL at Drigg in Cumbria. The main constraint related to the disposal of graphite as LLW is the carbon-14 content of the waste.

The remaining Drigg radiological capacity for carbon-14 is 1.5 TBq. This gives rise to a limit of 0.05 TBq per year, over the currently planned 30 year operating life.

Considering the total C-14 content of the graphite in Windscale Pile 1 is 6.9 TBq, then disposal of the Pile graphite as LLW would exceed the total radiological capacity of Drigg for C-14 by a factor of 4.6. This data demonstrates that it will not be possible to dispose of any of the redundant reactor graphite as low level waste.

The one exception to this is GLEEP. Because of the low levels of irradiation of the GLEEP graphite due to the low power output, the activity associated with the GLEEP graphite is much lower than in other graphite moderated reactors. The total C-14 inventory of GLEEP is approximately 8 GBq, this is not an insignificant quantity of C-14, but it should be acceptable for LLW disposal and this is the preferred route. Discussions are on going with the LLW disposal authority.

3.2. Incineration

A number of studies have been undertaken by UKAEA on the incineration of reactor graphite [4]. The incineration of graphite has a number of advantages in that it disposes of the stored Wigner energy problem completely, it greatly reduces the volume of waste i.e. 1400 m^3 of graphite could be reduced to as little as 35 m^3 of cemented ash product and filters.

The different possible methods of incineration are discussed below.

3.2.1. Conventional Burning

This process involves controlled combustion of the graphite. The graphite is first of all crushed into pieces of typical dimension 2.5 cm. The graphite is then placed in a furnace

where it is subjected to a blast of air at about 1000°C. This minimises CO production while allowing reasonably rapid boundary layer diffusion controlled combustion. Cooler air blasts would be needed to keep down the temperatures of the furnace walls and the graphite bed. Assuming that about 10 te of graphite could be incinerated per day, it would take over 600 days to dispose of all of UKAEA's graphite liabilities by this process.

The disadvantages of this process are the milling effort required, the production of active dust and the difficulties of the incinerator design.

3.2.2. Fluidised Bed Incineration

This process involves burning the graphite in a fluidised bed. The graphite would have to be ground to a powder (possibly down to 30 μm) to provide enough surface area for reaction with oxygen at incineration temperatures. The milling would produce dust that would have to be contained. This process could also lead to ignition if significant stored Wigner energy and air were present. These factors alone preclude this option on safety grounds.

Other disadvantages of this process include: the milling prior to burning produces waste itself, irradiated graphite can be very hard making milling more difficult and other materials can become mixed with the ash from the carbon thereby increasing the ash volume, as can any inefficiency during burning.

3.2.3. Power Laser Driven Incineration

An alternative method of incineration is by power laser. Advantages of this method are that no prior milling or crushing of the graphite is required before incineration. Thus the bricks can be loaded straight from the reactor core. It is claimed that the furnace design is very simple compared with earlier options. At the heart of the system is a 30 kW CO_2 laser. The laser beam heats the graphite surface to about 1500°C and rapid combustion takes place when O_2 is supplied. The laser itself can be outside the furnace area so does not require handling within an active area. Control is completely governed by the presence of the laser beam, as high temperatures are limited to one side of a single block. Any stored energy in the block is only a few percent of the total energy release and so does not constitute a problem. The suggested combustion rates are about 150 kg/h which with a 50% load factor would consume 700 te of graphite per year and take about 9 years to deal with the UKAEA graphite liabilities.

The main disadvantage of this process is that it is not proven technology on this scale and so would require a significant research and development programme to support this work.

3.2.4. Overview

The main disadvantages of incineration are the impact on the environment from gaseous emissions, both radiological and non-radiological, the generation of secondary wastes and the public perception of the acceptability of incineration of reactor graphite. Technical solutions could be used to reduce the emissions to the environment, but public perception and environmental pressure groups make this option unfavourable.

3.3. Disposal as ILW

UKAEA is working closely with the UK ILW disposal organisation, UK Nirex Ltd, to seek approval for the packaging and the disposal of UKAEA's graphite liabilities in their current form. In the absence of a finalised repository site, design or associated safety cases Nirex has developed a strategy which facilitates packaging of ILW by providing guidance through Waste Package Specifications [5], supported by the formal assessment of specific packaging proposals on a case by case basis. On completion of this formal assessment, Nirex is prepared to give advice on a packaging proposal and, where appropriate, provide a 'Letter of Comfort' to waste producers. Letters of Comfort provide assurances to waste producers that proposed packages are compatible with Nirex plans for the repository and associated transport system [6].

UKAEA has sought letters of comfort from Nirex for the WAGR moderator and reflector graphite, and the Windscale Pile 1 damaged and undamaged graphite. As most of the issues are common to all the wastes submissions for other reactor decommissioning wastes will be prepared closer to the time of decommissioning.

With the exception of WAGR the graphite will be packaged in Nirex standard containers either a 4 m ILW box, should the graphite waste meet the requirements of Low Specific Activity (LSA) or Surface Contaminated Objects (SCO) [7], or a 3 m^3 box. The waste may then be encapsulated in a cementitious matrix. However, discussions are on going with Nirex on proposals to leave certain graphite wastes unencapsulated because there is no requirement to encapsulate these wastes from a wasteform or pH buffering view point. If this route was adopted it is likely that the boxes would be packaged such that they could be grouted at a later date for void filling purposes should the future repository design require this.

The WAGR graphite will be packaged in standard density WAGR boxes. These are reinforced concrete boxes that meet the requirements of an IP-2 package.

4. TECHNICAL ISSUES ASSOCIATED WITH THE PACKAGING OF GRAPHITE FOR DISPOSAL

Having considered the options for the disposal of UKAEA's graphite liabilities there are a number of technical issues associated with the packaging of these wastes which need to be resolved. These technical issues are discussed in more detail below.

4.1. Wigner Energy

Wigner energy in irradiated graphite can be simply considered as follows. When a neutron hits a carbon atom in the graphite lattice, it pushes this carbon atom out of position. The carbon atom is not stable in this arrangement, and Wigner energy is the potential energy it has from being out of position. When the carbon atom has enough thermal energy i.e. it is hot enough, it is able to return to its position. Since the energy required to initiate a return to position is less than the stored energy, the excess is released as heat. Thus a finite amount of Wigner energy can be released by heating the piece of graphite above a certain temperature. If too much energy is released at one time or the heat is not carried away from the system a self-sustaining release can occur and the system will "run away with itself" i.e. get hotter and hotter even though no further heat is being put into the system.

It was thought that no Wigner energy was released up to a point 50°C above the temperature at which the graphite was irradiated, this could be as low as 80°C for parts of the Windscale Piles. This was known as the "start temperature". Now it is understood that Wigner energy is released at all temperatures, but it is released so slowly that it is difficult to measure and hence the "start temperature" is actually the point at which significant quantities are being released. Stored Wigner energy is especially a problem for graphite irradiated at low temperatures e.g. Piles, BEPO, GLEEP and the graphite reflectors of the MTRs. It is not such a problem for WAGR and the fast reactors because these were running at higher temperatures (above 100°C) where self-annealing of the Wigner energy from the low temperature irradiation will have occurred.

In the repository the temperature of the vaults is limited to 50°C, with excursions up to 80°C being acceptable for periods of up to 5 years. This is to limit processes within the waste such as corrosion and microbial degradation; it is also because the behaviour of the solubility of radionuclides is well understood within this temperature range. After emplacement of the packages in the repository and just before repository closure, the vaults will be backfilled with a special cementitious grout. This grout maintains a pH above 10 for an extended period, limiting the corrosion of the containers and the solubility of certain radionuclides especially uranium and plutonium. The integrity of this backfill is very important to the Nirex post-closure safety case.

When the packages are being backfilled, the temperature may increase to 80°C. Nirex are concerned that the levels of heat produced by the release of Wigner energy from graphite in waste packages could produce a run away effect that could damage the integrity of the backfill.

For the last two years UKAEA has been working with the Consortium, led by BNFL, tasked with decommissioning Pile 1, and with Nirex to try to demonstrate that the release of Wigner energy in the repository should not be of concern. The recent work has focused on establishing a fundamental understanding of the physics behind the release of Wigner energy. It has been postulated that the release of Wigner energy is a first order reaction, i.e. it is related directly to the number of "out of position" atoms. Therefore activation energies can be calculated for the movement of different carbon atoms back into their position in the lattice.

The output of this work has been used as an input into the thermal vault modelling being undertaken by Nirex. The results to date have led Nirex to conclude that the behaviour of Wigner energy is not adequately understood. There are too many uncertainties in the system (both repository location, depth, ambient temperature, design, as well as loading of the box) to give a firm statement that a box containing graphite with stored low irradiation temperature Wigner energy will not have a deleterious effect on the performance of the repository.

For Pile 1, the Consortium has chosen to go forward with plans to anneal the large blocks of graphite. They are still working with Nirex to try and make the technical case that slats and tiles, and graphite from the fire-affected zone can be disposed of without annealing.

For BEPO and the MTRs UKAEA are currently undertaking sampling campaigns and further calculation analysis to confirm the levels of Wigner energy associated with these graphite wastes. The data from these studies will then be input into the Nirex thermal vault models to determine whether annealing of the graphite will be required.

For GLEEP It has been calculated that the Wigner energy at the centre of the GLEEP core is approximately 2 cal or 8.4 joules per gram. It has been shown that, should all the Wigner energy in the GLEEP graphite be released simultaneously, the temperature of the graphite would increase by only 7°C. There are therefore no implications from the release of Wigner energy on the disposal of the GLEEP graphite.

4.2. Graphite Dust

The dismantling of the reactor graphite will generate graphite dust. Nirex waste package specifications require that all measures shall be taken to ensure that the activity in waste is effectively immobilised and loose particulate material is minimised. Nirex require graphite dust to be intimately immobilised to ensure that the package will perform in a predictable manner in an impact or fire accident.

Intimate immobilisation would be achieved by the encapsulation of graphite dust in some form of matrix. This would normally be a cementitious matrix. The concern with the cement grouting of graphite dust is that the dust does not "wet" easily and so some form of wetting agent is required to get large quantities of dust to combine with the encapsulation grout. This wetting agent and its degradation products would have to be acceptable to the disposal environment.

Where only small quantities of dust are present preliminary grouting trials at WAGR have demonstrated that the dust can be incorporated in the grout matrix. If a further layer of inactive grout is applied above the encapsulation grout as a cap then this will seal in any remaining graphite dust on the surface.

Several alternative methods for immobilising the dust are currently under consideration by the Windscale Pile 1 project these include:

- encapsulation of small quantities of dust in mixing bowls in the dismantling cell. These mixing bowls would then be disposed of in the final package;
- supercompaction of the dust in sacrificial cans, and supercompaction of the dust using a binder such as clay to provide a more stable product;
- the other solution is to encapsulate the dust in some form of polymer acceptable to the Nirex repository.

Work on the immobilisation of graphite dust is still at the early stages of development.

4.3. Graphite Flotation

Nirex require wasteforms to be fully immobilised in a cementitious monolith. However, the density of irradiated reactor graphite at 1.6 te/m^3 is less than the typical grout density of 1.8 te/m^3 hence flotation of the graphite can occur during grouting and in certain cases, depending on the loading of graphite in a package, this can be significant, even causing steel box furniture to float. This would result in a wasteform where the graphite was not fully immobilised by the grout.

The flotation of the graphite can be overcome by the use of an antiflotation device as part of the furniture inside the final package. This has to be designed to withstand the uplift forces caused by the graphite.

Floating of dust is a particular associated issue, and the processing will have to ensure that formation of dust from the graphite blocks is minimised and that grout capping of the grouted waste effectively immobilises small amounts on the surface.

4.4. Galvanic Corrosion

The integrity of a waste package during storage and after emplacement in the repository can be affected by corrosion of embedded metallic waste and box furniture causing wasteform expansion and gas generation. Higher corrosion rates could occur due to galvanic coupling caused by metal-to graphite contact. The currently proposed packaging strategy for graphite wastes, except for WAGR, is to encapsulate the graphite in Nirex standard packages manufactured from 316S11 stainless steel. In a number of instances the packaging of graphite will be associated with mild steel box furniture. WAGR graphite will be encapsulated in concrete boxes, the graphite will be held in mild steel baskets within the box [8].

UKAEA has undertaken studies to consider the effects of galvanic corrosion on the integrity of these disposal packages. Galvanic coupling between steel and graphite has been shown not to cause a significant increase in steel corrosion rates under the anaerobic conditions encountered in encapsulation grouts. Corrosion rates would be very low and similar to those for uncoupled steels. Higher corrosion rates may occur at an early age while wasteform conditions are aerobic. However, these rates will rapidly fall as oxygen is consumed by the corrosion process. Expansion from aerobic corrosion during this phase is estimated to be low; <250 microns. It has been concluded that corrosion of steels in the wasteforms considered so far will not threaten the waste package integrity either as consequence of expansive corrosion or gas pressurisation.

4.5. Radionuclide Inventory Determination

The radionuclide inventory of a package destined for disposal in a deep waste repository needs to be determined to comply with the requirements of the Nirex waste package specifications.

The radionuclide inventory of redundant reactor graphite is normally determined by sampling and analysis of different parts of the graphite to determine the radionuclide fingerprint. This fingerprint will then be applied to the graphite in association with some check measurement such as gamma spectrometry or dose measurements. The radionuclides of significance for the disposal of graphite in the ILW repository are H-3, C-14, Cl-36, Co-60, Sr-90, Cs-137 and Eu-154. Additional radionuclides may be significant for certain graphite wastes depending on the operational history of the reactor.

The irradiation of graphite in fast reactor systems may lead to the formation of certain key radionuclide not found in thermal reactor graphite. Work to determine the radionuclide inventory of graphite from fast reactors has still to be undertaken.

5. CONCLUSIONS

There are large quantities of graphite associated with a number of redundant nuclear reactors on UKAEA sites. In developing decommissioning and waste management strategies for these facilities, UKAEA have given detailed consideration to how to manage these liabilities.

Because of the carbon-14 content of the graphite only GLEEP graphite may be disposed of as LLW to Drigg. The remainder will require disposal as LLW or ILW in the proposed national Deep Waste Repository.

There are a number of technical issues associated with the disposal of graphite wastes. UKAEA is working closely with the ILW disposal company, Nirex, to develop waste management and packaging strategies for graphite waste which will ensure that waste is packaged in a stable and safe form for long term surface storage and ultimate disposal.

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