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Review of Treatment for Decommissioning Wastes

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HTR-N1 Review of Treatment for Decommissioning Wastes

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1 Introduction

This report for the review of treatments to wastes arising due to decommissioning, in response to HTR-N1 WP4 task 4.2.2, complements the report produced under task 4.1.1. The principal wastes arising during decommissioning are steel, concrete, and graphite. Other waste materials do occur, e.g. thermocouple materials, neutron sources, asbestos, densified wood, these are considered under miscellaneous, but their relative quantities are small.

Graphite wastes can occur during operations, particularly associated with refuelling where existing CO_2 reactors have graphite sleeves which become a waste stream, or for block type high temperature reactors, where blocks associated with the fuel are removed for refuelling. Such graphite wastes can be either ILW or LLW. The treatment of graphite wastes for decommissioning will usually be quite similar to those for operations, hence there is an element of commonality between this review and that for the operational wastes. Task 4.2.2 however, calls for a brief review of existing methods and procedures for dealing with steel and concrete from decommissioning. This report will therefore deal primarily with these wastes forms, first, and then as stated in the objectives review the state of art for dealing with graphite wastes.

Steel wastes can also occur during operations, as result of maintenance activities. Again such wastes can be ILW or LLW. The treatment for wastes arising under decommissioning will usually be similar to those for operations.

Concrete wastes, are on the other hand, unlikely to occur during operations, and therefore are likely to be exclusive to decommissioning. Concrete structures will usually have some form of shielding from the neutron fluxes of the reactor and are therefore LLW in nature, with a significant proportion being free release.

The treatment, or in most cases given the relatively few nuclear power station there have been decommissioned, the proposed treatment for each of these types of waste materials is reviewed below.

2 Steel wastes

Steel wastes can be either free release, LLW or ILW. LLW and ILW can be either activated or contaminated. The treatments for LLW and ILW will usually be similar, except that LLW wastes will be sent to Drigg, whilst ILW will be stored locally in containers awaiting the availability of the UK intermediate ILW repository. Free release steel, in UK definition meaning below 0.4 Bq/gm, is, after stringent measurement of activity, sold for scrap and can be used on the open market.

2.1 Activated steels

Activated steel will be either ILW or LLW. In either case, in the UK the first stage of treatment, according to size and shape, is likely to be size reduction to improve the packing factor, placed in a standard NIREX container, and grouted as described below under encapsulation. Prior to grouting, monitoring checks will be done to ensure that surface dose rates and weights are within regulations. A problem with steel wastes is that the weight restrictions may be reached long before the dose rate limits are approached, leading to low quantities of steel and high quantities of grout. To offset this problem, some mixing of wastes is allowed under the NIREX criteria. Steel wastes can be encapsulated in the same box as lighter materials, e.g. graphite or concrete. There are however certain rules to be observed in such mixed wastes, for instance there must be at least 10 cm of grout between the steel and graphite surfaces, to prevent the possibility of galvanic corrosion. If the conditioned wastes are likely to approach the dose rate limits, then the more active material will be placed in the middle of the container, with the less active material at the edge of the container, to take some advantage of self shielding.

LLW will be sent to BNFL's low level waste disposal facility at Drigg. Facilities exist at Drigg for grouting and compaction if this is necessary/desirable. Waste at Drigg is buried at near surface, in engineered trenches. The UK has no repository for ILW, therefore after treatment this category of waste will be held on site in a dedicated shielded store, until such time as the UK repository becomes available. The (ILW) decommissioning wastes from WAGR are held in a store local to the reactor building. The store has a design life of 50 years.

2.2 Contaminated steels

Contaminated steel wastes have been subject to a number of decontamination methods in the UK. These include:

High pressure water jetting
Basic mechanical decontamination
Heat treatment for release of tritium
Chemical decontamination
Shot blasting
Smelting
Strippable coatings

Examples of each are as follows

2.2.1 High pressure water jetting

Contaminated components of the Berkeley nuclear power station fuelling machine, which had been swept by the primary circuit CO_2 were subject to high pressure water jetting. A tented structure was erected over the components to be decontaminated and the water jet head. The tented enclosure was placed above the active drains for the site, allowing ready removal of the liquid arisings. The hyperbarric intensifier for the water jet equipment was placed outside the tented enclosure, some 30/40 metres away. The pressure used was 40,000 psi. The method proved successful in removing contamination, allowing components to be free released.

2.2.2 Mechanical methods

A trial decontamination of one of the Berkeley boilers started with mechanical decontamination (brushing and scrubbing, no further details available). The work was done in tented enclosure which was placed over the whole of the boiler, which had been lowered to the horizontal position at ground floor level. Following mechanical decontamination the boiler plate was treated for tritium release, see below. There are 16 boilers from the two reactors at Berkeley, and to date only one has been decontaminated.

2.2.3 Tritium release

Plate from the Berkeley boilers, following mechanical decontamination as described above, together with plate from some sections of the gas ducts, was treated for tritium release. The plate was first cut into sections approximately 1 m x 1 m and placed into an on site furnace. The metal was heated to 800°C to 1000°C to drive off tritium up the stack. The tritium was discharged to atmosphere, being within the allowable site discharge authorisations. After release of tritium in this manner, the plate was then free released.

It is not clear whether this practice would still be allowed today.

2.2.4 Chemical decontamination

A trial decontamination was carried out for the Windscale AGR (WAGR) boilers. A substantial part of the contamination in CO_2 cooled graphite reactors consists of carbon deposits embedded in oxide layers on the metal surfaces. Two type of oxide are grown during operations, basically ferric oxide in the hotter (upper) regions of the boiler and ferrous in the cooler (lower) regions. The different types of oxide require separate chemical agents, and therefore a number of different solvents were used. The trials were perhaps not successful, and eventually the remaining WAGR boilers were disposed as whole units to Drigg, where they were grouted on that site and disposed in purpose built trenches.

Chemical decontamination was also used extensively at the Capenhurst diffusion plant, and although this was mainly for aluminium materials, some steels were treated in this manner.

2.2.5 Shot blasting

Shot blasting can be used as a technique to remove surface decontamination from steels. A purpose built facility exists at UKAEA's site at Winfrith. Metals are transported there for decontamination, although it is not known whether this has yet applied to power station wastes.

2.2.6 Smelting

Smelting is not used widely in the UK and there is no equivalent of the Simpelkamp foundry in Germany for recycling contaminated steels as described above. A 16 ton capacity smelter exists at Capenhurst for the decontamination of aluminium, and a 2 ton smelter for iron and steels. The principle of smelting is that the contamination tends to concentrate in the slag, which can then be skimmed off and dealt with separately, say grouted in standard waste containers. The majority of the steel can then be cast into standard shapes, which greatly assist assay, and, if conditions are suitable, proved to be free release materials. On site smelters have been considered for use in the dismantling of UK power station reactors (not physically due to take place for another 70 years at least), but have not directly been part of the formal plans to date. The Japanese may deploy a smelter on site to assist in the dismantling of the Tokai Magnox reactor (due to be completed by about 2017).

2.2.7 Strippable coatings

Strippable coatings are an effective method of preventing contamination. The principle is that surfaces which will be subject to contamination are first sprayed or painted with the coating. At the end of operational life or at maintenance intervals, the coating is removed. The coating contains all the contamination, leaving the base metal as free release. According to application, the coating may have to be applied several times during plant lifetime. Coatings can only be applied to surfaces which are exposed to low level conditions of temperature, pressure, and contamination. Light duty ventilation ducts would be a good example of such applications.

3 Concrete wastes

As for steels, concrete can be either free release, LLW or ILW. LLW and ILW can be either activated or contaminated. The treatments for LLW and ILW will usually be similar, except that LLW waste will be sent to Drigg, whilst ILW will be stored locally in containers awaiting the availability of the UK intermediate ILW repository. A considerable amount of concrete wastes will be free release, and it will be necessary at sites to have effective monitors that can assay the concrete in bulk quantities.

3.1 Activated concrete

Activated concrete occurs particularly in prestressed concrete pressure vessels, where the innermost part of the concrete is separated from the core/neutron shield by only the steel liner, c 12 mm. Activated concrete can also occur in the bioshields of steel pressure vessels. The plan for treatment of such activated concrete in the UK would be as follows.

(i) Activation analysis will identify the divisions between the waste categories within the concrete (i.e. ILW areas, if any, LLW areas, free release). Sampling cores taken from the concrete will confirm the transition regions.

(ii) Dismantling cuts are made to remove the concrete in, say, cubes of 1 m^3 volume and in particular segregating the categories of waste.

(iii) The concrete cubes would then be placed in one of the standard waste containers for grouting, capping and lidding for subsequent disposal.

(iv) Because there are large volumes of concrete in the LLW category, and because it has low density (2.3 say) as described above, some of it may be placed in the same box as steel wastes, provided the NIREX rules are adhered to. The advantage is that the full weight allowance of the box can be taken advantage of, which might not be possible for steel wastes alone.

(v) When concrete is packed, a decision has to be taken on whether to remove the reinforcing bar, and in the case of prestressed concrete vessels whether to remove the prestress tendon conduits (the tendons themselves would have removed shortly after shut down). If concrete is packed in the same container as graphite for example, it is likely that the metal parts would have to be removed, to avoid potential galvanic corrosion activity with the graphite.

(vi) Concrete crushing may be used. Advantages of concrete crushing are that, activity high spots can be selectively removed, thus minimising the higher categories of waste, the packing factor in waste boxes is maximised, the metal parts can be better segregated, and there is better potential for mixing with the grout encapsulation.

3.2 Contaminated concrete

Contaminated concrete can occur in the bioshields of steel pressure vessels, and in all cooling ponds and in other active or waste handling buildings. In the UK, it was thought that the cooling pond walls, which had usually been lined before the start of operations, would be contaminated to a depth of a maximum of about 70 mm, somewhat deeper where cracks or construction joints occurred. Therefore the emphasis was on removal of a layer to this depth, most of the concrete being removed being LLW with some ILW. Initial methods of removing the layer of contamination were based on high pressure water jetting. The pressure used was about 40,000 psi, the water jet head comprising a spinning unit to remove a circular part of the concrete. The spinning jet head was traversed across the concrete to ensure even removal. This method was deployed remotely at both the ponds at the shut down Berkeley Magnox reactors and at the SGHWR at Winfrith. The technique can be used in air or underwater. When used in air, local containment is used, consisting of simple polythene tenting arrangements.

Since the early decontamination of these ponds, dry methods have come into favour. Tests have been carried out using remote operating vehicles, manufactured by Brokk, on which a scabbling head is mounted. The Brokk vehicle also carries a powerful suction hose leading directly to a waste disposal container. The concrete is removed in powder form and the vacuum system is very effective, almost none of the removed concrete escapes into the building (which itself would be contained and ventilated and filtered etc). The advantage of this method are the absence of wet wastes, and that the contaminated concrete is directly packed into the final disposal container for subsequent cementation, avoiding the need for intermediate handling.

Once the layer of decontamination is removed the remaining surface is assayed for any residual spots of activity, these are removed locally if necessary, leaving the bulk of the concrete structures free release.

The above methods of decontamination can be said to have progressed beyond theory and research and development and to have been put into practice. A comprehensive range of concrete decontamination methods can be considered to be as follows:

1. High Pressure Water Jetting (in practice)
2. Dry Mechanical Scabbling (in practice)

3. Microwave Scabbling
4. Laser Scabbling
5. Electrical Heating of Reinforcing Bars
6. Concrete Shaving
7. Dry Ice Jetting
8. Insoluble Wet Abrasive Jetting
9. Chemical Leaking
10. (Diamond Wire Sawing)
11. (Drilling and Bursting)
12. Crushing

Methods 3, 4 and 5 have been the subject of research and development by the Japanese for the decommissioning of their experimental reactor JPDR, and to some extent in the UK. A description of the techniques is given below.

Microwave Scabbling

Concrete absorbs microwave energy within a depth of 10 to 30 mm of the surface. The water in the concrete matrix converts into steam and causes rapid expansion. This principle has been used to remove the surface layers of activated concrete.

A prototype microwave concrete surface remover was developed by the Japan Atomic Energy Research Institute. Three 5 kW magnetron units were used to generate 2450 MHz microwaves which were directed into a flat concrete surface by a moving irradiation head. Experiments were carried out to determine the depth of concrete removal against rate of travel of the system. Concrete debris and dust were removed simultaneously by attaching a piped vacuum system to the rear of the irradiation head.

A second set of experiments were carried out in Japan using a microwave output of 30 kW at 915 MHz. The concrete began to disintegrate within 2 to 5 minutes of irradiation and the internal concrete surface temperature reached 150°C. Concrete material was removed up to 60 mm into the surface and the irradiation head moving speeds varied from 100 to 200 mm/minute.

The limitations of the method might be the difficulty of removing concrete beyond the first layer of reinforcement.

Electrical heating of reinforcing bars

Experiments were first carried out in Japan on various forms of electrical heating of internal reinforcements in concrete structures with a view to removing the surface layers. This work has lead to a wide expansion of full-scale trials on reinforced concrete specimens and sections of the existing structures. Electric currents of 1000 to 3000A at 50 Hz were applied directly to the reinforcing bars in these trials and the concrete surface material was successfully exfoliated with a minimum level of associated concrete dust and fine particles.

An alternative method of applying heat to the reinforcing steel by inductive methods have also been investigated in Japan. A rage of power inputs, 100 to 200 kW and frequencies of 3-200 KHz were tried in the first experiments.

This method of electrical heating is particularly intended for the removal of surface layers of concrete. It has the potential advantage that men and equipment need not necessarily enter the area involved, until follow on operations become necessary. Despite this obvious advantage the method has not been put into widespread use.

Laser beams

A series of experiments were undertaken in Japan to study the feasibility of using lasers to cut concrete sections. A high output CO_2 laser system was developed with specific features to enhance the cutting effect, prevent the fumes generated from interrupting the laser and enable steel reinforcement to be cut. Experiments were conducted with a wide range of laser outputs from 5 to 15 kW, at cutting speeds of 25 to 300 mm/minute. It was found that concrete cuts up to 180 mm deep could be achieved with a power output of 15 kW and a cutting speed of 25 mm/minute.

Concrete shaving

Concrete shaving has been achieved by the use of diamond impregnated grinding discs. The discs require to be traversed across the concrete surfaces. The reaction forces are substantial, probably requiring mechanical equipment rather than hand held tools (a possibility and particularly if remote deployment is necessary is the use of Brokk remote operating vehicles). Considerable quantities of dust are generated, and direct contact between the grinding wheel and the concrete contaminates the former, leading to some secondary waste arisings.

Dry ice jetting

In this method, CO_2 liquid or solid pellets are fed through to a jet nozzle, which is traversed across the concrete surfaces. The pellets are formed by a cold unit, compressor and dryer unit. It is to be considered at the research and development stage only, the author not being aware of practical experience. The CO_2 produced is an obvious respiratory hazard to operators, and operator visibility can be difficult. The stand off distance is reported not to be critical. An advantage is that there is very little secondary waste.

Insoluble wet abrasive jetting

In some ways this method is similar to water jetting. Abrasive material such as metal powder is sprayed onto the concrete surfaces and is traversed over the walls and floors. Disadvantages include generation of secondary waste, difficulties with operator visibility and likely airborne contamination.

Chemical leaching methods

This method uses a chemical reagent, either acidic or alkaline to leach radionuclides from the concrete. The reagent is applied by spraying or jetting, or in the case of small concrete volumes by immersion in a bath of the reagent. The method has potential to leach the contamination from surface layers of the concrete, leaving the bulk concrete behind, hopefully at free release levels. A major disadvantage would be the considerable quantities of secondary waste created.

Diamond wire saving

The technique is well known and is primarily associated with cutting concrete rather than decontamination. It has become popular since its use on Fort St. Vrain to remove sections of the pre-stressed concrete pressure vessel. This cutting method could be used in conjunction with some of the above decontamination methods, to assist in the process.

Drilling and bursting

Drilling and bursting is also a well proven method of cutting/separating concrete. As for diamond wire sawing it could be used in conjunction with a number of the above decontamination processes.

Concrete crushing

Where decontamination has not reduced the concrete to powder or small particle size, concrete crushing may be used to facilitate, packaging or grouting prior to final disposal. Concrete crushing may also help in the detritiation of concrete, and in the identification of waste categories and segregation.

Detritiation of concrete

One of the major problems thought to be present in contaminated concrete, are quantities of tritium. Research and Development is currently planned in the UK on heat treatment (and possible other methods) to detritiate the concrete. The tritium might simply be released up a stack if within discharge limits. As stated above, crushing the concrete first is likely to assist the detritiation process.

Comparison of Concrete Decontamination Methods

Method	Advantages	Disadvantages	Comments
High Pressure Waste Jetting	Proven Methodology Good rates of surface removal. Water can be recirculated. Can work	Large quantities of wet wastes, even if water recirculated. Poor Operator visibility. Potential for airborne contamination	Was used in the early days, but appears to be loosing favour

Method	Advantages	Disadvantages	Comments
	underwater. Stand off distance not critical.		
Dry Mechanical Scabbling	Proven technology, and is being further developed. Associated vacuum systems virtually eliminate airborne contamination and can route concrete dust and rubble direct to waste drums/containers	High reaction forces required. Scabbling cutters may need to be changed periodically. Corners are a problem.	Becoming the favoured option with UK regulator and utilities.
Microwave Scabbling	Acceptable removal rate can remove up to 100 mm depth. Secondary waste arisings are minimal. Tolerance towards variable stand off distances.	Operators need shielding from microwaves. Success rate after first pass is limited. Potential for airborne contamination	Still at R + D stage.
Laser Scabbling	Relatively easy to apply.	Localised high energy and heat input, concrete rubble can be ejected. Potential for airborne contamination. Difficult to operator to see application. Depth of removal limited to, say 10 mm per pass.	Some laser techniques will require gas services, e.g. CO_2, He, N_2. A few applications have been noted otherwise at R + D stage.
Electrical Heating of Reinforcing Bars	Men and equipment do not have to be deployed with contaminated buildings. Therefore secondary wastes can be limited. Amounts of	High electrical currents and energy input required. Reinforcing bars may be difficult to locate.	Still at R + D stage.

Method	Advantages	Disadvantages	Comments
	concrete dust report to be minimal.		
Concrete Shaving	Where contaminated concrete occurs in more than one waste category (e.g. LLW free release, the method is good for segregation).	Operations produce noise and vibration. Potential for airborne contamination. Operator visibility can be a problem. Grinding wheels need replacement. High reaction force required.	The technique requires little R + D. Equipment such as rigid platform structures, or Brokk vehicles are required for deployment.
Dry Ice Jetting	Process is basically commercially available. Stand off distance is not critical. Secondary wastes are minimal.	Good ventilation required to avoid CO_2 respiratory hazards. Operator visibility can be impaired.	Equipment can be considered as general purpose, with use for, say, steel wastes, as well.
Insoluble Wet Abrasive Jetting	Basic equipment and abrasives commercially available. Stand off distance not critical.	Potential for airborne contamination. Difficulties with operator visibility. Abrasives will add to secondary wastes.	Methods not in general use.
Chemical Leaching	Potential to remove contamination, leaving free release level concrete intact. Chemical agents commercially available.	High quantity of secondary waste for treatment of chemicals.	Still at the R + D stage.

4 Graphite

Actual experience of treatment and disposal of graphite from shut down reactors is limited. Of the graphite reactors that have been shut down world wide, Fort St. Vrain is the only reactor of significant size that has been fully decommissioned, with graphite sent to disposal facilities. Windscale AGR, (WAGR), in the UK has (at the time writing, mid 2002) started to treat graphite from dismantling operations of the core and AVR in Germany will do so in the next few years. Graphite reactors benefit from a prolonged period of safestore, and for this reason few have started decommissioning. Hence, disposal of graphite tends to be based either on research and development and theory rather than practice, or on the work done for operational wastes in the case of graphite sleeves (UK, France, Spain, and Japan all have this type of waste). Therefore this report examines the international thinking for the treatment of graphite wastes and covers the following.

(i) the range of options for the disposal of irradiated graphite (including any pre-treatment)

(ii) assessment of technical disposal criteria

(iii) disposal methods being proposed internationally (UK, France, Japan, etc)

(iv) identification of future activities and research and development required to support graphite disposal.

4.1 Range of options for disposal of graphite

Worldwide, graphite reactors exist in the UK, France, Germany, Japan, USA, Russia and the former states of the Soviet Union, Italy, Spain, China and other countries. A wide range of ideas and options for disposal of graphite have been proposed and these are identified below.

Some of these options may, or in certain cases have to be, preceded by pre-treatment, and this subject is described in later.

4.1.1 Raw storage

This option is the first and most obvious. Graphite is retrieved from the reactor, or vaults in the case of sleeves or other operational waste, dried where necessary, and put into storage canisters, waste boxes or drums. The advantage of raw storage is that most of the other options for disposal are left open, which can be selected after further research has identified the optimum. The disadvantage is that the storage period is likely to be temporary only, until a final storage solution is available, implying double handling/transport of raw graphite. Since this might involve large quantities, it may not be regarded favourable by regulators. Therefore, storage may well be limited to the locality of the source. Treatment in Germany appears to be a hybrid situation, whereby graphite is stored in raw form for 30 years, prior to ultimate disposal. The current HTR-N1 R&D proposals to release the volatiles from the graphite at the start of the storage period, and therefore ease eventual disposal, are therefore particularly relevant to this option.

Safestore may be considered as an important part of raw storage. The UK currently has the longest safestore proposals, planning to leave reactor cores intact for 85 years, (although the economic justification is based on 70 years). Research and development has been undertaken in the UK on the ability of the graphite to withstand this long period of safestore. Research items particularly considered were the effects on long term atmospheric corrosion, and potential radiolytic corrosion (due to nitric acid formation). Apart from decommissioning taking place in conditions of greatly reduced dose rates, the waste management advantage of safestore is that some graphite can actually be reclassified from ILW to LLW during safestore period.

4.1.2 Sea dumping

All countries belonging to the European Community have accepted a moratorium on sea dumping, effectively since 1984. The UK accepts that this moratorium could be indefinite, but reserves the right to continue research and development and to re-open debate if the political climate were to change. In reality, it is more likely that a worldwide ban will be accepted by all countries.

At the time of moratorium, both the governments of the UK and France believed that sea disposal was probably the best practical environmental option (BPEO) for disposal of all but high level wastes, including graphite. The graphite wastes would be encapsulated and containerised prior to disposal in accordance with requirements. Both polymer and cement encapsulation have been accepted for sea disposal. Where such waste is disposed in deep water the dilution factor would be very large, and even this would not take place for many years, after breaching of the containment and slow leaching outwards. The resulting additional radioactivity is likely to be undetectable against background levels.

4.1.3 Volume reduction techniques

None of the above disposal techniques have included volume reduction processes. The following methods are, in part of whole, concerned with volume reduction of graphite. They are:

(i) mechanical methods

(ii) densification

(iii) incineration

(iv) pyrolysis.

4.1.3.1 Mechanical methods

The proposals for removing the graphite core from the Tokai Magnox power station start with a mast manipulator scheme capable of extracting seven bricks at a time. When the bricks have been removed to the first part of the waste route, some of the bricks are then cut in the longitudinal direction so that the pieces so formed can be inserted into the channel bores of the other bricks. Using this method the packing factor in a typical container would be increased from 35% to 70%.

Dust is usually a problem with graphite reactors, irradiated graphite produces more dust than unirradiated graphite. The cutting process is therefore to be undertaken in a glovebox type enclosure, with dust removal. Once the graphite bricks have been cut, an encapsulation process is intended, using cement mortar. Graphite dust arising will be vacuumed off and also encapsulated in cement mortar.

4.1.3.2 Densification

Densification is a process whereby graphite is compacted to a level approaching the maximum theoretical density – thereby eliminating pores and voids, as well as machined bores. Densification can be achieved by electrolytic or chemical means. This is an emerging technology and its effectiveness is not yet certain.

4.1.4 Incineration

Next to encapsulation, incineration is possibly the most popular method for disposal of graphite. The advantages of incineration are very volume reduction combined with gradual and controlled dilution of the ∃ emitters (^{3}H and ^{14}C) in the atmosphere. The final ash residue can then be stored on a surface site without difficulty. Incineration releases to the atmosphere represent 70% if the radioactivity, but only 1% of the radiotoxicity. There are several different methods for incineration and these include:

(i) fluidised bed

(ii) laser incineration process

(iii) induction process

(iv) plasma incineration.

Much of the research and development for the fluidised bed (and other incineration methods) have been carried out in France, and the following is a description of their work.

(a) Fluidised bed incineration

Graphite blocks that make up the reactors cores or spent fuel sleeves are placed in a crushing installation with hammer-type and cylindrical crushers. The objective is to achieve a final average grain size of 1 mm, without too large a fraction of particles of less than 100 :m size, so as to limit the risks of dissemination.

In either case, the finely crushed graphite is then fed into a dense or circulating fluidised bed type of combustor, whose features include a high fluidisation air flow rate and high turbulence in the combustion region. The fluidised bed consists of powdered refractory material. Solids are separated from the combustion gases by a cyclone separator, and recycled via a recirculation loop, which contains only non mechanical parts.

The combustion gases leaving the recirculation cyclone separator have a low concentration of dust. This dust contains fly ash, fine refractory particles arising from slight wear of the fluidised bed, as well as a small quantity of unburned graphite. Incineration of the graphite is completed in a post-combustion chamber.

A program to validate this process, supported by CEA and EDF, was carried out using a prototype incinerator installed at Le Creusot, France. The validation program included 22 – 12 h tests and one 120 h test, and was constructed around four phases:

(a) search for the stable operating points

(b) determining the sensitivity of the parameters and the limiting values

(c) determining incinerator behaviour during transients and incidents, and

(d) simulating industrial operation (the 120 h test).

The results were very satisfactory. Combustion is complete and perfectly controlled.

Safety aspects were also studied. These included:

An assessment of Wigner energy release during the crushing of the graphite, leading to the adoption of slow crushing methods.

An assessment of Wigner energy release during the incineration process. The conclusion was that the combustion temperature would be increased by a small value, and could be easily controlled by the process and equipment.

An assessment of the effects of graphite dust, and the avoidance of explosible conditions at any stage.

Note that the above method of incineration, and indeed any incineration method, could be significantly enhanced by the introduction of appropriate catalysts. Lead compounds, particularly lead oxides are known to increase greatly the oxidation rates for graphite, and it is possible that their introduction could obviate the need to fine crushing.

(b) Ash residue

The remaining waste, i.e. the ash residue, would form about 1% to 2% of the original volume. The intention would be to encapsulate these ashes. Much of the radioactivity would be concentrated in the ash, therefore in order to comply with storage and transport requirements it might be necessary to ‘dilute’ the ash. This in turn would limit the volume reduction effect.

Three types of embedding materials were tested in the CEA Laboratories for the fluidised bed incineration ash:

(i) cement

(ii) epoxy resin

(iii) mixed cement – epoxy resin matrix.

Glass and ceramic matrices were also investigated, but not so thoroughly. Tests on stability, mechanical strength, sensitivity to composition, leaching, and irradiation were validated. On the basis of the results obtained, the volume reduction (evaluated at 20) and financial considerations, the mixed cement-resin matrix was selected.

(c) Laser incineration

A laser incineration process was developed by the CEA of France in the 1990s. Only laboratory tests were performed, and this process is neither qualified nor available at an industrial scale.

(d) Induction

An induction process was also developed in the 90s by the CEA. It consists of heating graphite pieces by induction, under pure O_2 gas flow, so as to gradually consume the graphite pieces. The development stage of this process is similar to the laser incinerator as above.

(e) Environmental impact of incineration

Environmental impact assessments for the incineration of graphite have been carried out both in the UK and France. The usual assumptions are that one commercial core is incinerated each year. The following assessment, for the case of incineration at Marcoule is typical.

The atmospheric concentration at ground level resulting from release at stack level depends on the atmospheric transfer phenomena. The dispersion coefficient depends on the stack height (100 m was considered), wind speed, diffusion conditions and distance from the point of release. The selected data correspond to the conditions in Marcoule (France).

Dose calculations were performed for a person working on the site (dispersion coefficient = 1.2×10^{-7} sm^{-3}) and for a member of the general public living near the site (dispersion coefficient = 3.6×10^{-7} sm^{-3}), on the basis of the mass radioactivity concentration worked out from data and calculations.

In the case of aerosols, the efficiency of the filters was evaluated at 10^4, (i.e. 99.99%). For the sake of prudence, a factor of 10^3 (i.e. 99.9%) was used for ruthenium and caesium. For gases (tritium, carbon, chlorine), filters were considered to be inefficient.

For carbon (as CO_2) and tritium (as tritiated water vapour), specific radioactivity conservation in the biosphere has been considered. For chlorides and aerosols, the releases lead to a deposit on vegetables and on the soil. The deposit on plant is submitted to biological and physical elimination, computed as an exponential decrease.

Considering the ultimate waste (embedded ash), the $\forall$ emitter concentration level allowed the analysis of shallow land burial for these residues.

Three pathways were taken into account; inhalation of gas and aerosols, external exposure from the deposit on the ground and in the case of members of the general public, ingestion of contaminated vegetables. The tritium release leads also to contamination by absorption of tritiated water vapour through the skin.

The results of this assessment for a 800 t/year incineration throughput led to the conclusion that the atmospheric release would induce a maximum dose of 0.1 mSv/y due to ^{14}C by ingestion pathway. The effects of other radionuclides would be trivial.

The conclusion of this assessment was that it was feasible to recover and incinerate contaminated graphite wastes in a safe manner, since it would induce a maximum annual dose of 0.1 mSv/y. This dose could be considerably reduced if the emission is planned when crops are not growing.

Residue disposal would never induce an annual dose higher than 1.5×10^{-3} mSv/y.

(f) Incineration with effluent release to the sea

A variation to the above, studied in France, is to scrub and wash down the off gas effluents, so as to transform them into a contaminated liquid to be released to the sea. Four sites of release were investigated, with a dilution factor varying from 2.7×10^{-3} to 2.7×10^{-4}. The critical groups considered were fishermen and tourists. The predominant nuclide was ^{14}C through animals ingestion pathways. In both cases the annual dose was low. However, in view of the progressive restrictions being imposed on releases to the sea, it is unlikely that this line of research will be pursued further.

(g) Conversion to grout

Since release of ^{14}C to atmosphere is the principal objection to the incineration of graphite, alternative means of capturing this isotope have been considered. One idea is to pass the CO_2 arising from incineration through a calcium hydroxide $Ca(OH)_2$ bed, capturing the ^{14}C as calcium carbonate. The calcium carbonate would then be used, in say, a deep repository as lime rich grout, put in as backfill that is considered necessary anyway. Such an approach, if both feasible and allowed by the regulatory authorities, would avoid ^{14}C release and also lead to significant effective volume reduction.

(h) Choice of location for incineration

One of the decisions to be made in respect of incineration, is whether to burn the graphite in situ, at the site of origin, or whether to retrieve the graphite, and after suitable transport, burn at a central location. The problem with the former is that individual sites many not be suitable for conversion to incineration; the problem with the latter is that retrieval of graphite, from say reactor cores, followed by subsequent packaging suitable for transport, undermines any economic advantage that may be gained from incineration.

(i) Conclusions for incineration

France (and Japan) have examined the possibilities for incineration. In the case of France, a pilot plant was built for the research of the fluidised bed incineration. Tests were carried out on unirradiated graphite. Incineration is considered as a good solution, possibly more economic than encapsulation for large scale use. In real terms, there would be a low impact on population and environment. However, for public perception reasons, and possibly for ALARA compliance, sub-surface disposal of graphite is the preferred option in the UK, France, USA and Japan. Notwithstanding the above, recent reports from Japan indicate that incineration of the graphite sleeves from the shut down Tokai reactor is being considered.

(j) Pyrolysis

Pyrolysis is a process whereby steam is passed through organic materials, in conjunction with low levels of oxygen. The reaction products are volatile gases and a fixed carbon char. Under certain conditions, the char may then be further treated with steam to gasify the carbon content in the char.

In a simplified form, the process has been used in the USA and Canada for treatment of spent ion resins containing organic or carbonaceous matter. The Studsvik Inc has patented the Thermal Organic Reduction (THOR) process which utilises pyrolysis and steam reforming technology. An initial plant for processing a variety of low level wastes has commenced in the USA, such wastes including graphite. Volume reductions quoted are similar to those for incineration, (~80:1). It is claimed that there are advantages over incineration; that it can be carried out in a better controlled containment and that loss of the radioactive materials in the off gas system is much reduced or eliminated. The process can be used to separate the carbon in the graphite from other radioactive elements in the moderator, thereby facilitating the subsequent treatment of each type of waste.

Studsvik claim that the process could be used to dispose of graphite in situ, within the pressure vessel. This would be done slowly, using very dilute steam in an inert atmosphere as a means of removing the graphite from the reactor core in a slow and controlled manner. Before accepting this claim, NNC would like to be better convinced of the capabilities of the process. As with incineration there is the possibility of capturing ^{14}C in the CO_2 off gas with a CaOH solution, using the carbonate formed as backfill material in a repository.

(k) Transmutation

Usually, transmutation has been seen as a treatment for minor actinides. Its use for irradiated graphite has been considered in Japan and the UK at least. It is not clear to NNC how such a process could be utilised.

(l) Recycling

Irradiated metals have already been decontaminated and recycled through the use of the melting technique and purpose built foundries.

For graphite, it is unlikely that direct recycling (i.e. the use of graphite blocks in another nuclear facility) will be possible on a widespread basis. The main idea for graphite, is to use it in cast iron containers for storage or transport of nuclear materials, or as shielding material. Metals recycling is carried out in Germany on an industrial scale in the Simpelkamp foundry. (The Simpelkamp nuclear smelter has the capability to accept radioactive scrap-up to a certain limit, smelt, deal with the active off-gases arising, and produce cast items. The nuclear foundry exists alongside, but physically segregated from, a conventional foundry. It is not known whether the Simpelkamp facility can yet accept graphite and recycle in this manner).

4.1.5 Pre-treatment considerations

4.1.5.1 Drying

The first and most obvious pre-treatment that may be necessary is the removal of water that may have accumulated in storage, either in air or underwater. Depending on the encapsulation process chosen, the drying could consist of simple draining of the graphite over a period of time, or may involve a small amount of heating or dry air methods. Water removal would be particularly desirable for some encapsulation process including the main cement option.

4.1.5.2 Wigner energy

Wigner energy accumulation (‘stored energy’) occurs in graphite under neutron irradiation because atoms are displaced from their normal lattice positions into configurations of higher potential energy. Some simultaneous thermal and irradiation annealing takes place, but there is a nett energy gain which is a function both of irradiation time and temperature. The higher the irradiation temperature, the lower the amount of ‘stored’ energy. Fuel-element sleeves therefore generally contain the lowest amounts of Wigner energy. In all cases, a saturation point may be achieved in terms of the total amount of stored energy for long periods of irradiation.

A proportion of the stored energy can be released if the graphite is heated to about 50°K above its irradiation temperature, although a temperature in excess of 2000°K is required before all the energy can be released. In extreme cases, with graphite originally irradiated at low temperatures (between ambient and 100°C, say) an initial temperature increase can release sufficient energy to result in self-heating to high temperature.

Wigner energy has been assessed as an issue for graphite reactors, since experience at the UKs Windscale Piles. These reactors, which were air cooled, operated at low temperatures, which led to considerable amounts of stored energy. This stored energy was periodically released by raising the graphite above a critical temperature, which would cause some of the graphite to undergo a self-propagating release. The heat released would spread to other parts of the core. Temperatures of over 350°C were reached. It was during one of these annealing periods that the famous Windscale incident of 1957 took place, when the release of the Wigner energy caused some fuel elements to catch fire.

For the UK Magnox and AGR reactors, there will be negligible amounts of Wigner energy and so it is not seen as an issue for the disposal of this graphite. (The minimum inlet gas temperature for a Magnox station would be about 150°C which is higher than the gas outlet temperature for the Piles.)

It requires to be ascertained therefore, what temperatures will have to be reached during the treatment options, to be certain that an inadvertent release of Wigner energy will not occur, or will not result in significant events. The maximum temperature likely to be reached during the curing of cement encapsulation, for example, is not likely to exceed 150°C. Subsequent storage conditions, both long term or intermediate are not likely to exceed this figure either. Note that the incineration or pyrolysis options raise the temperature of the graphite above the release level, and therefore are considered as self annealing.

4.1.6 Assessment of technical disposal criteria

This part of the report is presented in table form, identifying in the first column individual technical (including in some cases cost and possible social factors) criteria that have been found relevant in other countries and international comparison, and in the second column the relevance of the criteria to different countries. The table is produced for ready understanding and to promote dialogue/debate.

Technical disposal criteria	Relevance
1. Definition, and number of waste categories	According to definition of categories, graphite (and other waste) may have to be conditioned differently, and sent to different repositories
2. Cost of final disposal for each waste category	Determines whether decontamination or volume reduction is cost effective
3. Total quantities of graphite in the reactors	Can influence policies
4. Initial impurities present in graphite	Determines radioactivity of nuclides after irradiation. Main nuclides are ^{14}C, ^{3}H, ^{36}Cl, ^{60}Co, ^{41}Ca, ^{55}Fe, ^{59}Ni, ^{65}Zn, ^{152}Eu, ^{154}Eu
5. Irradiation history	Lifetime reactor fluence will determine the radioactive inventory of graphite. Criteria 3, 4 and 5 enable this to be calculated, at final shutdown and at any time thereafter
6. Presence of certain nuclides	May effect disposal to certain repositories and require waste level reclassification to higher categories
7. Graphite sampling programme	Radioactive inventory calculations are based on idealised situation. Sampling can identify departures from theory
8. Waste assay	Prior to emplacement in containers (particularly in the case of encapsulation) it will be necessary to ensure the surface dose rates etc are acceptable

Technical disposal criteria	Relevance
9. Safestore period	A long safestore period, in situ allows decay of the shorter lived nuclides. Most significantly, it allows reactor structures to be dismantled more easily
10. Minimum operating temperature for graphite	If any part of the graphite is irradiated below, say, 150°C, then that part of the graphite should be considered for annealing of Wigner Energy during treatment and conditioning
11. Type and size of disposal container	May determine volume reduction techniques. In case of some types of encapsulation, may influence ability to produce a homogeneous structure (e.g. into pores)
12. Type and nature of interim and final disposal repositories	Could affect the preferred option for treatment
13. Ability to accept radioactive backfill in repositories	Allows ^{14}C to be captured as carbonate, in say the incineration and pyrolysis disposal options
14. Public perception and ALARA criteria on release of ^{14}C	Affects ability to accept incineration, (or pyrolysis) options without ^{14}C capture
15. Leaching rates for irradiated graphite	Affects assessment of long term storage conditions
16. Quantity of, and particle size of dust generated during operations or dismantling	Dust may need to be controlled during graphite retrieval to prevent explosibility. Graphite dust may need more careful treatment and conditioning
17. Criteria for encapsulation - mechanical properties mechanical stability thermal stability resistance to internal radiation	 a homogeneous structure is required without voidage reasonable stability during the encapsulation process is required must be capable of withstanding freeze and thaw cycles if in near surface disposal in cold climate. For transport requirements may have to withstand high temperatures (UK typical could be 800°C for 30 min) cumulative affects of the internal radiation to be demonstrated by accelerated tests
18. Sea dumping	Not currently available for UK and EU countries. Otherwise considered as the best practical environment option by both UK and France

Technical disposal criteria	Relevance
19. Facilities available for recycling, particularly melting and casting, and attitude of regulator	Possibility exists to recycle some waste graphite in shielded containers for example. Requires facilities similar to Germany's Simpelkamp foundry or others

4.1.7 Methods being proposed internationally

4.1.7.1 United Kingdom

The United Kingdom has 40 commercial graphite reactors, 26 Magnox reactors and 14 advanced gas cooled reactors. The total mass of graphite in the cores of these reactors is about 75,000 t. Ten of the reactors, two each at Berkeley, Hunterston A, Trawsfynydd, Bradwell and Hinkley A are shutdown, and are being decommissioned. Windscale Pile 1 is being decommissioned, but Pile 2 will be placed in Safestore.

Experimental graphite reactors have all been shutdown. These exist at Windscale (WAGR), at Harwell (BEPO and GLEEP), and at Winfrith (DRAGON) and at various universities.

Within the UK, the electricity utilities have a policy of long term safestore for the graphite reactors. The reactor internals will not be dismantled for 85 years. The reactors will be subject to a defined Care and Maintenance regime during these years. The advantage of such a delay is that radioactive decay will greatly simplify the dismantling, allowing limited personnel access to the workface. However, some graphite will have to be disposed of, at least for temporary storage (i.e. graphite blocks from WAGR and Pile 1, Magnox sleeves, and AGR sleeves).

WAGR is being decommissioned early to demonstrate the techniques to be used on the larger gas reactors (and also to demonstrate to the UK public that decommissioning is feasible). Approximately 210 t of core graphite will be retrieved, passed through the sentencing cell, and sent through the waste route. The waste route, as mentioned earlier, will encapsulate the graphite in cement and pulverised fuel ash mortar. The graphite will be encapsulated in 4 m^3 concrete boxes and either transferred to a nearby intermediate store in the case of ILW or sent to Drigg in the case of LLW. The intermediate store has a design life of 50 years. It is worth noting that all waste from WAGR will be treated in this manner, because of the way that the waste route has been set up. By 2003 all the graphite from the core is programmed to be dealt with.

The shutdown Magnox reactors at Berkeley and Hunterston A, both had forms of sleeved fuel. The graphite sleeves have been stored in bunkers on the sites. Such graphite, along with other ILW, is currently being retrieved from the Berkeley bunkers. It will be disposed of, for interim storage, in the on site ILW store.

The United Kingdom Government is currently re-assessing its policy for final ILW disposal. The current select committee thinking is that there should be an interim period of storage in a near surface, shallow repository, prior to final storage deep underground. The emphasis is on gaining public acceptance of the principal, and more importantly the location of the store. It is not expected that even the interim store will be available for several years.

All practices and procedures will ultimately require the endorsement of the Nuclear Installations Inspectorate and the Environment Agency. It is not cleat that the Nuclear Installations Inspectorate will accept an 85 year delay before all reactors are dismantled. (The original safestore period of 120 to 130 years has already had to be changed.) It is possible therefore that some reactors will have to be dismantled, and graphite cores disposed of much earlier.

As a final note, although banned by the EU for the foreseeable future, both the UK and France deemed sea dumping to be the best practical environmental option.

4.1.7.2 France

France has shutdown graphite reactors at Marcoule (G1, G2, G3), at Chinon (A1, A2, A3), at St. Laurent (A1, A2) and at Bugey 1. The total mass of graphite in the core and reflector of all these reactors is ~18,500 t. All the cores and reflectors are still in place. Although not formally declared, the idea is to have a 40 year safestore period, which if adopted, would result in graphite retrieval from G1 Marcoule in 2008 and Chinon A1 in 2013, these being the earliest reactors to be shutdown.

Chinon A1, A2, St. Laurent 1, 2 and Bugey 1 have graphite sleeves arising from spent fuel during operations. Most of the graphite sleeves from Bugey 1 have been conditioned and disposed of at the La Hague disposal facility. Unlike the sleeves at Chinon and St. Laurent, the Bugey 1 sleeves were without stainless steel wires. The Bugey sleeves were treated as follows:

- the sleeves were first drained (to remove absorbed water)
- then put into concrete boxes, details of which were 2.11 m x 1.56 m x 1.29 m, wall thickness 15.19 cm, weight (empty) 4.55 t
- 32 sleeves (580 kg) in each box
- 1370 t of graphite were sent to the La Hague disposal facility where they were grouted and disposed of.

The La Hague facility is now full and graphite waste containing ^{14}C cannot be disposed of at the 'Centre de L' Aube' facility as it is not licensed for this waste.

The last graphite sleeves (a total of 194 t) remain at the Bugey site. A further 4070 t of graphite sleeves from Chinon and St. Laurent, together with their stainless steel wires, are in temporary storage. It is likely that any future disposal option will require separation of the stainless steel wires, to allow for ^{60}C decay.

The lack of disposal facilities in France for graphite has led to research and development on incineration, as described earlier in the text. France, together with Japan, and in part, the UK, consider incineration to be the most economic disposal method currently available. However, the authors observe that French research into incineration is effectively halted. Incineration is not accepted by the public, even if it complied with ALARA principles. It is probable that French graphite will initially be disposed of (temporarily at least) in a surface disposal facility. A facility close to the existing Centre de L' Aube is being considered.

4.1.7.3 Germany

Germany has two main graphite reactors, the HTRs at Hamm-Uentrop and Julich. Graphite cores remain in place awaiting final storage decisions. Research is on going for the fuel pebbles which are mostly graphite. Research is proposed under this EC contract, HTR-N1, into the heat treatment of graphite, to determine the effect and feasibility of driving off the volatiles in the interim storage period, prior to final disposal. It is worth noting that the Simpelkamp foundry has the ability to recycle (low level) metal scrap, and could possible be used to recycle a small quantity of radioactive graphite in cast containers.

4.1.7.4 Spain

Spain has one shutdown commercial graphite reactor at Vandellos (which is essentially a repeat of the French St. Laurent design), and one experimental graphite plant. Vandellos has 2,500 tons of graphite core and reflector and 1000 tons of sleeve graphite. The sleeves from the Vandellos vaults have been retrieved, and have been separated from their nimonic springs.

Spanish researchers were considering an electrolytic coating process for the Vandellos graphite, prior to a land disposal option.

4.1.7.5 Italy

Italy has one graphite reactor, Latina, a Magnox station with approximately 2000 t of core and reflector. This was shutdown in 1987. Initially a 40 year safestore period had been proposed, but more recently a 20/25 year safestore has been implied by government policy. NNC is not aware of any research and development in Italy for graphite disposal.

4.1.7.6 Japan

Japan has one shutdown graphite reactor at Tokai, (which was closed in 1998), and one experimental HTR, which started up in 1999. There are 1,600 t of core and reflector graphite at Tokai, and a considerable quantity of graphite sleeves arising from 32 years of operation.

Japan has examined a range of options for graphite disposal, including incineration, and the theoretical aspects of transmutation. However, the plans for the Tokai graphite are as follows:

- retrieval from the reactor, seven bricks at a time by use of purpose built remote machines

- volume reduction by sawing in glove box type containment, with suitable extraction of dust.

- encapsulation in cement mortar.

4.1.7.7 USA

The USA has graphite reactors at Hanford, the high temperature reactor at Fort St. Vrain, and the X10 reactor at Oak Ridge National Laboratory (ORNL) and many other facilities. The reactor at ORNL has been retained as a National Historic Landmark.

At Fort St. Vrain, the HTR has been decommissioned almost to 'greenfield' status. Graphite core blocks were retrieved from the reactor underwater. The graphite retrieved was in two categories. Graphite bricks not containing fuel elements were, with some exceptions, considered as low level waste. These bricks have been transported to the Hanford site, in Richland Washington. They were placed in ¼ in. (6 mm) thick stainless steel containers and buried in trenches 45 ft. (~14 m) deep.

The graphite containers were stacked from the 45 ft. to the 8 ft. (~2.5 m) level, then covered with top soil for the last 8 ft. Graphite containing fuel blocks have either been retained on site or at the Idaho Chemical Processing Plant (Idaho National Engineering and Environmental Laboratory) until US national disposal facilities are available. The fuel blocks from the Peach Bottom cores are also in storage at Idaho.

In the 1980s, there was a plan to build a federal repository for graphite at the Hanford site, primarily to store graphite from reactors used for research and defence purposes. However, this plan has been put on hold due to concern of ground water contamination due to ^{36}Cl, which has a half life of 300,000 years.

^{36}Cl results from the irradiation of ^{35}Cl which is present in the graphite as an impurity resulting form the purification process. At the present time all graphite from the defence reactors has been put in SAFSTOR status at the Hanford site until a decision has been made for the final repository.

4.1.7.8 China

China appears to be investigating sites for land repositories being, like Russia, endowed with vast tracts of sparsely populated land. A demonstration land-disposal repository facility in the remote Lanzhou nuclear facility has been proposed in which graphite components will be disposed of. It is inferred, but not explicitly stated, that this could lead to a similar disposal route for the graphite from early plutonium-producing reactors. The demonstration site is in a very remote area and this is advantageous from the point of view of political acceptability, although the area suffers from high wind erosion. Geological and ground-water features are considered to be ideal. 54 disposal 'cells' are located in nine reinforced-concrete channels; armoured concrete drums are described as the principal containers.

Decommissioning requirements are already being taken into consideration for the graphite from the new HTR-10 reactor which is now starting operations, but no final decision has been reached about the preferred route.

4.1.8 Advantages and disadvantages of various types of treatment and disposal

Table 1 Advantages and disadvantages of different disposal options

Option	Advantage	Disadvantage
Raw storage – temporary or as safestore	- retains options for disposal - found to be acceptable for UK, 120 years - safestore gains advantage of radioactive decay - raw disposal used in USA for low level waste	- may require double handling in case of interim storage - may be restricted to local storage - problem with graphite dust - may have problems with contamination
Encapsulation	- actually used in France for sleeve disposal - subject to much research and development in UK - preferred UK method - can be economical	- does not reduce volume
Sea dumping	- seen by UK and France as best practical environmental option - experience (of other wastes) prior to 1984	- banned in EU and elsewhere for foreseeable future
Densification	- volume reduction, 20-40%?	- process not sufficiently researched
Incineration	- major volume reduction, $\simeq$ 70.1 - ash residue easy to encapsulate - seen by France and Japan as most economical, after sea dumping - safe release of Wigner energy (subject to controlled crushing)	- not acceptable to public in France, Japan - may require capture of ^{14}C - may not be possible in-situ, therefore transport required to central facility - throughput limited to one reactor per year?
Pyrolysis	- volume reduction as incineration, $\simeq$ 70:1 - residue easy to handle	- process is emerging, performance details for graphite unknown - may require capture of ^{14}C as for incineration

Table 1 (cont'd)

Option	Advantage	Disadvantage
	- safe release of Wigner energy - claims to be capable of in-situ (or adjacent facility) disposal - process proven for spent ion resins	
Transmutation	- being studied in Japan and UK at least - otherwise no advantages seen	- all theory, no practice yet - major application is for minor actinides rather than graphite
Recycling	- being considered by a number of countries - proven for low level steel waste	- requires purpose built facility for recycling as cast iron - not able to treat all graphite wastes?

Table 2 Advantages and disadvantages of different encapsulation methods

Encapsulation method	Advantage	Disadvantage
Cement (usually mixed with blast furnace slag or pulverised fly ash)	- simple, well proven process - low temperature process - economic - requires only reasonably dry graphite - good fire resistance - good strength properties - radiation stability - good self shielding - high pH matrix - self supporting matrix - good ageing properties	- low initial strength prior to setting - may set prematurely blocking plant/pipes - some permeability
Polymer	- relatively simple, well proven process - low temperature process - high ability to withstand strains - low permeability - self supporting matrix	- possible fire hazard - more expensive than cement - process plant requires periodic solvent flushing - requires graphite to be dried - less radiation stability than cement

Table 2 (cont'd)

	- good for sea dumping	- may be more susceptible to leaching than cement
Bitumen	- known process - no need to dry graphite - good leaching properties - reasonably economic	- fire hazard - high temperature process - low radiation stability - limited self shielding - abandoned by France after R&D, and may be abandoned by Japan after waste accident
Resin sand	- reasonably simple process - resistance - high strength matrix - good self shielding	- potential fire hazard - high temperature process - requires graphite to be Dried - high porosity - possible low leaching Resistance - less radiation stability than Cement
Vitrification	- used for other wastes, HLW in UK and France - fire resistant - high radiation stability - good self shielding	- very expensive - high temperature process - process requires off gas Facilities - probably requires graphite to be dried - process requires fine control to avoid cracking, and poor leaching properties
Others	- either no advantages or too little know about process - further details from NNC on request	- several - further details from NNC on request

5 Miscellaneous wastes

The following is a resume of miscellaneous wastes material types, not consisting wholly of graphite, steel, or concrete.

5.1 Asbestos

Asbestos was used widely in the UK nuclear industry in the 1950s and through to the mid/late 1960s. It was used to insulate the external surfaces of steel reactor pressure vessels and boiler shells. A quantity of asbestos insulation has been removed from the boiler shells of the shut down Berkeley Magnox station. Removal was effected by operators working manually in air suits. Once removed, the asbestos is placed in small sections, and double bagged in polythene. The asbestos is disposed of at licensed sites, whether it is above the free release limit or not. In any event, it is unlikely that any of the asbestos would be above LLW limits.

5.2 Neutron sources

Neutron sources are used in the UK to facilitate start up of reactors, allowing the neutronics instrumentation to pick up readings at low power levels. The neutron sources usually consist of a beryllium and antimony inner section, contained within a stainless steel can. Disposal soon after shut down would be difficult. The plan is to allow these sources to decay in situ during the UK's planned long term safestore, and then to dispose of them in the usual manner i.e. cement grout in approved containers, followed by the disposal in the UK's repository, when available.

5.3 Thermocouples

In the UK strategy, the majority of thermocouples will remain in the reactor during the safestore period. The plan would be to retrieve them during decommissioning of the reactor. They will be encapsulated in cement grout, as for all the other wastes. One possibility, to effect volume reduction is to place the thermocouples inside the channel bores of the graphite bricks prior to grouting.

6 Encapsulation

(Note this part of the report is similar to the report on operational wastes, several types of wastes being common to both operational and decommissioning aspects.)

Although very little of the world's irradiated graphite has been finally disposed of yet, encapsulated is currently seen as the most favoured method. There are different forms of encapsulation proposed:

(i) cement

(ii) polymer

(iii) bitumen

(iv) resin sand

(v) vitrification

(vi) stone/ceramic

(vii) metallic combination.

Significant research has been undertaken in the UK and France for some of the above, and particularly for the cement option. As mentioned in section 2.1 above, the UK decommissioning proposal for the commercial graphite reactors is long term safestore, so the emphasis for actual disposal is on the (prototype) Windscale AGR (WAGR), Windscale Pile No. 1 and graphite sleeves. There are two types of sleeves – those from AGR fuel elements stored at BNFL's Sellafield site, and those from certain Magnox stations, where the graphite is stored in vaults on site.

For any encapsulation process, basis problems to be overcome are:

(i) floatation of graphite, particularly smaller particles and dust, to the top of the grout

(ii) lack of penetration of the grout into the interspaces, and to a lesser extent the pores of the graphite.

A two stage encapsulation process may be used to overcome floatation problems. In the initial stage the grout or encapsulation medium covers the waste, in a drum or box, up to a few inches of the final surface. After the initial grout has set, a capping layer is then introduced. The two stage process ensure that particles are kept down. A separate lid can then be put on or cast into the drum or box.

To ensure full penetration of the encapsulation medium the concentration of grout feed, or encapsulation mix has to be controlled and the use of an appropriate plasticiser is essential. This is particularly the case for waste in smaller containers such as drums, where sacrificial paddles may be used to assist in the mixing process. The size of container may also affect mixing abilities. In the UK there may be a need to consider drums at 200 P and 500 P sizes, and 3 m^3 and 4 m boxes.

It is logical to carry out tests on unirradiated graphite first, and indeed this has been done in the UK and France for encapsulation. It is important to realise the relevant differences between irradiated and unirradiated graphite in this respect. Irradiated graphite contains greater porosity. It can therefore absorb more water. Irradiated AGR graphite in the UK has been reported as absorbing 10% of its weight of water, after long periods of storage under water. For other forms of graphite, such as PGA, which have lower initial density, and therefore higher porosity, absorbed water content after storage under water could be even higher. Encapsulation trials may therefore have to be carried out on both soaked and dry graphite, or alternatively the graphite may have to be dried out.

For encapsulation, research and development knowledge of the following factors are required:

(i) heat output from radioactive decay

(ii) radiation dose to the encapsulation matrix

(iii) heat released, and temperatures reached during the setting process.

In order to complete the assessment, it would also be necessary to know and model the conditions of storage in the repository, e.g. layout and density of containers, backfill medium used.

Encapsulation trials would provide data on the following properties:

(i) mechanical properties – homogeneous matrix

(ii) mechanical stability

(iii) shrinkage/expansion testing

(iv) impact testing

(v) resistance to radiation damage

(vi) leaching rates.

For the mechanical properties, it will be necessary to demonstrate that all the waste has been encapsulated. As a complementary issue, it will also be necessary to demonstrate that there is minimal voidage in the matrix.

The prime consideration for mechanical stability is that the matrix will have sufficient strength to withstand handling and transportation to the intermediate or final repository. The strength should also be sufficient to withstand accident conditions during transport, such as dropped loads. Moreover, this strength should ideally be achieved within a short time duration after setting. Once settled, the matrix should remain structurally and chemically stable throughout the life of storage and disposal.

Certain encapsulation processes, including cement, will be subject to dimensional changes resulting from hydration during curing. These changes should be monitored so that they may be compared with other mature structures of a similar kind, and which are known to be stable, and therefore give confidence that the waste matrix will remain stable.

With regard to thermal stability, it will first of all be necessary to consider whether there will be interim storage at or near to ground level. If this is the case, then according to location, the effect of winter freezing will have to be demonstrated. The effects of freeze and subsequent thaw will have to be determined, and the number of cycles for which this will happen. The freezing cycles should be demonstrate that there are no adverse effects on the mechanical stability.

High temperature stability will also be required. The greatest temperatures conceivable will be from fire hazard. The actual temperature reached will depend on the accident scenario assumed. In the UK, reasonable assumption would be 800°C for 30 min. This is the accident scenario to be assumed during transport of nuclear fuels.

An assessment of the specific heat and thermal conductivity of the matrix will enable heat build up from radioactive decay to be derived. For reasonable conditions of underground storage the heat build up is not expected to be significant.

Radioactive decay will take place in the body of the matrix, both in any interim and final storage. The cumulative effects of the irradiation on the stability of the structure will require to be demonstrated. This could be assessed by subjecting samples to high levels of irradiation in order to simulate the long term effects of self irradiation.

Finally, and equally applicable to any of the disposal techniques, the concentration of waste material needs to be calculated to ensure that surface dose rates from the container satisfy regulatory needs.

6.1 Cement encapsulation

Of all of the encapsulation matrices researched, in both the UK and France, cement has emerged as the preferred option. It is proven technology, fire resistance and does not produce high temperatures in the curing/setting period and is economic. It provides a high strength matrix (after curing), radiation stability, self shielding, and the high pH value will be an advantage in any final deep repository.

Possibly the first irradiated graphite to be encapsulated in the UK will be from WAGR. The waste packing route at this site has been set up and is in use, albeit for components other than graphite. The WAGR graphite cores will be packaged in shielded concrete boxes, with Ordinary Portland Cement (OPC) and Pulverised Fly Ash (PFA) grout/matrix. The PFA in the grout and matrix offers very good protection against subsequent fire hazards. The concrete boxes themselves are 4 m^3, although they are not standard NIREX boxes.

6.2 Polymer encapsulation

Spent ion resins at the UK Trawsfynydd power station have been encapsulated in polymer in a custom built plant at the site. The chemical materials for the polymer matrix are supplied by the Dow Company. Although the process was originally intended for use with sea dumping, the plant has been recently been recommissioned, and the polymer encapsulation has been accepted by NIREX for sub-surface disposal. The major advantages of polymer encapsulation are that it is a simple process, to proven technology, in a low temperature process. It provides a good strength matrix and offers low permeability. The disadvantages are (compared to cement) that is relatively expensive, is a possible fire hazard and has reduced radiation resistance.

6.3 Bitumen encapsulation

Bitumen encapsulation has been research in the UK and particularly in France. It has good leaching resistance and can accommodation any initial water present. Another advantage of bitumen in the high ratio of graphite to matrix (4 parts graphite to 1 part bitumen), thus ensuring relative waste volume minimisation. Although the process was reached extensively in France, it has never been fully utilised there, and is now abandoned in that country.

The bitumen process has been used in Japan for waste treatment, although not necessarily for graphite. The obvious disadvantage with bitumen is the potential for fire hazards, and indeed at the PNC (now JNC) waste treatment works at Tokai, Japan, a bitumen explosion/fire did occur in 1997. There was also a fire reported at the Karlsrhue research facility in Germany.

6.4 Resin sand

Resin sand matrices have been considered by a number of countries. It is no longer considered as a preferred option. There are doubts that the sand grains can penetrate through the matrix and the graphite pores. Also the encapsulation process is complicated and involves high temperatures.

6.5 Vitrification

Vitrification, or glass encapsulation is being used for high level wastes. However, it is not thought suitable for the volume requirements of graphite for low and intermediate level wastes. The process requires fine control and involves high temperature processing, which if the process is not sufficiently controlled, can lead to cracks in the matrix structure due to differential thermal expansion. In turn the cracks can offer leach paths from the waste. The process has matured in recent years for HLW.

6.6 Other encapsulation media

Other encapsulation media that have been considered include:

(i) stone/ceramic

(ii) metallic combination.

Stone/ceramic require high temperature and pressures. Metal alloy with low melting points are expensive. It is considered that these last two options would require extensive development.

Finally, it should be noted that combinations of the above encapsulation techniques have been proposed. Lithuania, for example, has considered the study of ceramic-glass encapsulation for graphite, covered by an outer coating of polymer for shock absorption.

7 Future activities and R&D

NNC believes that there are possible advantages in understanding further some of the emerging disposal technique. In particular these are:

1	Pyrolysis	What are the advantages of this method over incineration? Could it be made publicly acceptable. What is the real feasibility of setting up the process in-situ for reactors? Can the C_{14} be adequately captured as carbonate and then used as grout backfill in a repository.
2	The coating process	More details on the process reported for treatment of the Vandellos sleeve.
3	Transmutation and recycling	It will be interesting to monitor how these ideas develop in the next few years.