Wigner Energy in Irradiated Graphite and Post-Closure Safety

R&D Technical Report P3—80/TR

Publishing Organisation
Environment Agency
Rio House
Waterside Drive
Aztec West
Almondsbury
Bristol BS32 4UD
Tel: 01454 624400 Fax: 01454 624409

ISBN 1 85705 898 4

Dissemination Status
Internal: Released to Regions
External: Released to Public Domain

Statement of Use
This document is intended for reference and use by the Environment Agency in its statutory role of regulating discharges from nuclear licensed sites.

Keywords
Wigner, energy, graphite, waste, disposal, management

Research Contractor
John Rees
Serco Assurance
424.4 Harwell
Oxon OX11 0QJ
Tel: 01235 432996 Fax: 01235 433169

Agency Project Manager
Martin Gilbert,
Ghyll Mount, Gillan Way,
Penrith 40 Business Park, Penrith, CA11 9BP
Tel: 07879434626 Fax: 01768 865606

Further copies of this report are available from:
Environment Agency R&D Dissemination Centre, c/o
WRc, Frankland Road, Swindon, Wilts SN5 8YF

tel: 01793-865000 fax: 01793-514562 e-mail: publications@wrcplc.co.uk

EXECUTIVE SUMMARY

Background

Wigner energy is formed in graphite as a result of irradiation with neutrons in research, production and power reactors. If sufficient activation energy is supplied subsequently, the Wigner energy can be released, potentially in an uncontrolled way. This is thought to have been connected with the fire of 1957 in one of the Windscale Piles.

The temperatures in commercial power reactors are high enough to release a significant fraction of the Wigner energy during reactor operation. There are, however, a number of research and production reactors in the UK that operated at low temperatures. Most of these facilities are at various stages of decommissioning, and this step will generate graphite wastes containing Wigner energy that may be released at relatively low temperatures, with the solid potentially becoming self-heating. Current sources of such graphite at Sellafield include Pile 1 and a mixed waste silo in which Magnox, graphite and aluminium are the main components.

The overall aim of the study reported in this paper was to provide the Environment Agency with a basis for developing guidance to waste producers on the management of irradiated graphite wastes. Current policy notes that the preferred route for managing the wastes in the short term has to accommodate the requirement for safety in the long term, that is, the route should be sustainable. Important themes associated with this include not foreclosing options if the current policy of geological disposal changes as a result of the current review and the ease of reworking the waste packages under these circumstances.

Objectives

The objectives of the study were as follows:

- To review the literature on Wigner energy relevant to waste management.
- To identify the options available for managing irradiated graphite wastes.
- To review experimental and modelling techniques for assaying Wigner energy and its release, and the confidence that can be placed in them.
- To determine the events that could lead to a release of Wigner energy in a repository.
- To understand the impacts this could have on the post-closure safety of a repository.
- To define key criteria for comparing options for managing irradiated graphite wastes.
- To make recommendations to stakeholders concerning unresolved issues.

Results

The main routes discussed in the literature for managing irradiated graphite are:

- Packaging of the untreated waste, followed by storage and disposal ('direct disposal').
- Annealing of the graphite, followed by packaging, storage and disposal.

- Incineration, followed by packaging, storage and disposal of the residue and secondary wastes.
- Steam reforming, again with packaging, storage and disposal of the residue and secondary wastes.

Direct disposal of graphite could lead to some of the stored energy being released after final closure of a repository. Sufficient input heat may be provided by the curing of the backfill; radiolytic decay; and the heat released during corrosion of metals and attack by microbes on organic wastes. The results of published modelling suggest that if the package is wholly occupied with graphite, a large rise in temperature could occur that could on present evidence lead to a significant decrease in the safety of the repository. The same work suggests that if half the volume of the package is occupied by cement grout, there may be a large enough heat sink to restrict the rise in temperature to a level that would not degrade repository safety. However, because of lack of data, it has not proved possible to date to complete the validation of these models, and so these results should be interpreted cautiously. In this study, a methodology based on the use of risk matrices and performance indicators has been developed to understand the impact of temperature rises on repository safety. For the mixed graphite waste, it is unclear at present whether the concentrations of irradiated graphite are sufficient to pose a potential problem in storage and disposal.

Annealing of graphite at 200-250°C is a reasonably well understood process that could in principle produce graphite that can be loaded into containers at the 100% level. Such a package may be suitable for geological disposal and could be reworked. However, residual levels of Wigner energy at this lower temperature peak have still to be determined. Incineration and steam reforming have the advantage that the graphite is destroyed in the short term. The volume of secondary wastes due to trapping of tritium, carbon-14 and particulates may, however, exceed that of the original graphite if it is decided not to release them to the atmosphere.

Recommendations

Overall, a framework has been established for relating actions taken in the short term to manage graphite wastes to environmental issues arising at all timescales, including the long term. In this context, a number of criteria have been developed that it is recommended be included in determinations of the Best Practicable Environmental Option for managing individual irradiated graphite wastes.

Other recommendations cover process uncertainties (discharges to the environment, the effectiveness of annealing, and system costs); modelling temperature rises in repositories (reviewing the status of model validation and further assessments); and impacts on disposal safety (safety margins with regard to temperature rise in a repository, identification of initiating events, and a methodology for assessing the risks associated with Wigner energy release). It is also recommended that publication of work on managing graphite wastes and its independent review should continue to help build confidence in the route(s) eventually selected.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

1. INTRODUCTION

1.1 Background

The irradiation with neutrons of graphite in research, production and power reactors leads to the displacement of some of the atoms within the crystal lattice. The positions in which the displaced atoms are held are of higher energy than the original graphite, and this potential energy is called Wigner energy. The carbon atoms concerned can migrate to form defect clusters, and the Wigner energy can be released from these groupings if sufficient activation energy is provided. The release can be rapid and lead to substantial increases in temperature.

In commercial power reactors (Magnox and Advanced Gas-cooled Reactors (AGRs)), operating temperatures are sufficiently high that significant release of Wigner energy occurs during reactor operation (see subsection 2.1 and Figure 2.4). This is known as auto-annealing. In reactors operating at relatively low temperatures, Wigner energy accumulates in graphite, and a deliberate annealing operation is needed to release a component of it. The remaining Wigner energy in both commercial reactors and low-temperature facilities requires relatively high annealing temperatures for its release (see subsection 2.1), temperatures that are unlikely to be encountered during the management of the waste under normal conditions. If Wigner energy were to be released before repository closure (that is, during packaging, storage or transport), the quality of the packages could be degraded. However, this type of behaviour is likely to be outside the regulatory requirement for passive safety, as discussed later in this Section.

The Piles at Windscale are examples of reactors that operated at sufficiently low temperatures for the graphite to accumulate Wigner energy, a circumstance that may have contributed to the Pile 1 fire in 1957 (Marsden *et al.* 1997). Pile 2 was also closed as a result of this incident. Consideration is currently being given as to how the core of Pile 1 should be decommissioned, including establishing a management route for the waste irradiated graphite. Irradiated graphite articles are also currently stored on the Sellafield site as part of a mixed solid waste, and a management route is being sought for them as well. This waste contains potentially reactive metals (uranium, Magnox and aluminium) as well organic wastes that contain cellulose, and are therefore degradable by microbes (Department of the Environment, Transport and the Regions and Nirex, 1999).

Lower-volume irradiated graphite wastes that contain Wigner energy derive from operation of British Experimental Pile O at Harwell and materials testing reactors at Harwell and Dounreay. Levels of Wigner energy and radioactivity in graphite from the Graphite Low Energy Experimental Pile at Harwell are sufficiently low that the material is acceptable for disposal as low-level waste (LLW) at Drigg (Wise 2000). All the other graphite wastes discussed in this Section are classified as intermediate-level wastes (ILW), based on the inventory of radionuclides (Wise 2000). The levels of the long-lived radionuclide ^{14}C and other isotopes make the material unacceptable for disposal at Drigg.

In relation to the Wigner energy content of these wastes, if the energy were to be released during one of the stages of the management route:

- waste treatment;
- storage on the surface;
- transport to a final repository;
- emplacement in an operating repository; and
- the post-closure period of a sealed repository,

a range of hazards could be realised, including fire, release of radioactive materials, and, in the post-closure period of a repository, degradation of the long-term safety of the facility.

Regulation of the stages of the waste package life cycle is carried out by the Environment Agency and Her Majesty's Nuclear Installations Inspectorate (HMNII). Co-ordination of regulatory activities is ensured through a Memorandum of Understanding between the two organisations (HSE and Environment Agency, 1996). HMNII is concerned with the prevention of accidents and the protection of the workforce and public, while the Agency is concerned with the effects on the public and the environment arising from any materials discharged from sites as gaseous, liquid or solid substances. This includes the final disposal of the wastes as well as discharges to rivers, the sea and the atmosphere at earlier stages in the waste package life cycle. Approval for authorising disposal of wastes lies with the Agency. The main focus of the work reported here is the methodology for evaluating proposals for managing wastes containing irradiated graphite, so that their treatment in the short term is sustainable in terms of long-term safety in an engineered repository.

The overall aim of this study is to provide the Agency with a basis for developing and maintaining a thorough understanding of the management of irradiated graphite wastes, particularly the implications for their long-term management. The management route selected for the waste in the short term has to accommodate the requirement for safety in the long term, so that the management route selected is sustainable. In addition, the route chosen needs to meet the following requirements of current policy (Review of Radioactive Waste Management Policy, Cm 2919, 1995):

- It should demonstrably represent the Best Practicable Environmental Option (BPEO) for managing the wastes.
- The wastes should be treated at an early stage so that they are placed in a chemically and physically stable form, with safety then being achieved by passive means. Emphasis has been placed on this aspect in the last few years by the failure to date to establish a site for a deep repository in the UK, with the consequence that wastes will have to be stored on the surface for an extended period.
- It should not foreclose future options for the long-term management of the waste, for example, if prolonged storage on the surface is ultimately selected (House of Lords Select Committee on Science and Technology, 1999).
- Reworking of the packages should be practicable so as to accommodate unforeseen deterioration or changes to the conditions of final management.

The current activities of decommissioning of the core of Pile 1 at Sellafield and defining a management route for the mixed waste containing irradiated graphite are driven by the need to achieve passive safety in surface storage of the wastes concerned (Marsden *et al.* 1997). The structure of the damaged reactor is believed to be unsound, for example with respect to a seismic event. In contrast, Pile 2 was not involved in the fire of 1957, and is therefore structurally more sound. Its decommissioning is not considered an issue for the short term.

1.2 Objectives of this Study

Within this overall aim, the detailed objectives of the study are:

- To review the open literature on Wigner energy (available at January 2001) that is relevant to radioactive waste management.
- To identify the options available for treating and packaging the wastes.
- To review experimental and modelling techniques for assaying Wigner energy and its release, and the confidence that can be placed in them.
- To determine in a systematic way the events in a repository that could lead to the release of Wigner energy and prioritise them.
- To understand in a qualitative sense the impact that these priority events could have on safety in the post-closure period.
- To determine the criteria that should be used to compare options.
- To use the outputs summarised above to characterise the options (as far as this is possible).
- To use this experience to define the principles and a framework for assessing proposals for treating and packaging irradiated graphite wastes. This effectively creates a methodology for establishing BPEO.
- To make recommendations to stakeholders for exploring currently unresolved issues.

1.3 Structure of the Report

The literature review forms Section 2 of this report and covers experimental assay of Wigner energy and its modelling; the degree of confidence that can be placed in these methods; options for managing graphite wastes; strategies for emplacing waste packages; conditions that could lead to Wigner energy release in a repository; and the identification of currently unresolved issues.

Section 3 examines assay techniques for Wigner energy in terms of the options identified for managing graphite wastes. Sections 4 and 5 are concerned with Wigner energy release in a repository: respectively the identification of events that could lead to this and its effect on repository safety. Section 6 uses the findings of earlier Sections to identify the key criteria that need to be taken into account when comparing options for managing graphite wastes and establishing BPEO. The conclusions and recommendations of the study are to be found at Section 7.

2. LITERATURE REVIEW

This Section contains the literature review described in subsection 1.3. Publications on the fundamental aspects of the generation of Wigner energy, its release, and related modelling and experimental techniques are relatively common, and include books as well as papers. However, there is significantly less available that is specific to issues in the management of irradiated graphite wastes. This applies both to options for managing the wastes, the release of Wigner energy in a repository, and the effects of this on repository safety. The main source of papers concerned with these topics is a conference organised by the British Nuclear Energy Society and the International Atomic Energy Agency on Characterisation, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Power Reactors that was held in Manchester on 18-22 October 1999. Papers from this meeting have appeared in the journal Nuclear Energy and have been published by the IAEA as a CD-ROM in the TEC-DOC series. The references are shown in their Nuclear Energy form where available. Further work is in progress, particularly in the field of radioactive waste management and graphite wastes, and the open literature will continue to evolve beyond the cut-off date for this study of January 2001.

2.1 Wigner Energy and its Release

Most of the work on Wigner energy was performed in the 1950's and early 60's and was primarily concerned with the operation of the Windscale Piles in the UK. It was not considered appropriate to review all the Wigner energy reports issued during this period, as much of the information contained in these reports has already been incorporated into the standard textbooks on graphite such as Nightingale (1962) and Simmons (1965) and to a lesser extent in Kelly (1981) and Reynolds (1968). This review draws extensively on the information contained in these books and only references the basic reports where necessary.

In 1942 (Fermi 1942) it was realised that moderating fast neutrons using graphite would displace the carbon atoms and Eugene Paul Wigner proposed that this process would store energy within the graphite and that this energy would be released if the graphite was heated. Subsequently, the excess energy possessed by the interstitials and its release during thermal annealing became known as 'Wigner energy' or simply 'stored energy'.

Each time a neutron collides with a carbon atom in graphite it transfers a fraction of its energy to the atom, until eventually the neutron becomes thermalised, i.e. it has the same energy as the thermal motion of the atoms (~0.025 eV at room temperature). The binding energy of the carbon atoms in graphite ranges between 12 and 60 eV, and the collision of a fast neutron (possessing an energy of several MeV) with a carbon atom transfers sufficient energy not only to displace this primary atom (a primary knock-on), but also for this atom (if it has sufficient energy) to displace other atoms, producing a cascade of displaced atoms (Figure 2.1).

Many of these displaced atoms have sufficient thermal mobility to find their way back into vacant sites (vacancies) within the graphite lattice, but some become trapped between the lattice planes where they are known as interstitials (Figure 2.2). Interstitials have a higher potential energy (~ 2.5 eV according to Iwata, Fujita, and Suzuki 1961) than the atoms in the graphite lattice. As the temperature of the graphite increases the interstitials become more mobile, increasing the probability that they will recombine with vacancies and release their excess potential energy as heat. Interstitials may remain trapped between the layer planes as single atoms or they may form complexes of two or more atoms depending on their mobility, and hence temperature (see Figure 2.2).

The formation of interstitials and the accumulation of Wigner energy are a strong function of irradiation temperature (Figure 2.3). If graphite is irradiated in liquid nitrogen (-196°C) and then subsequently heated, energy is released in the form of three "peaks" centred approximately at -80°C, +200°C and +1200°C (Figure 2.4). The origins of these peaks are still not completely understood and there are various interpretations (e.g. Hennig and Hove 1956, Reynolds and Goggin 1960, Iwata and Suzuki 1963). The interpretation due to Iwata and Suzuki is that below -80°C the interstitials exist as single atoms or loose 'atomic clusters'; at -80°C these loose clusters start to form 'molecular clusters' consisting of pairs of carbon atoms (C_2 clusters). At 200°C these C_2 clusters start to condense into larger clusters and dislocations, and at 1200°C, vacancies start to become mobile and migrate, destroying the interstitial clusters and dislocations. Electron microscope studies (Reynolds, Thrower and Sheldon 1961, Reynolds and Thrower 1963) have shown that at 1550°C the number of irradiation-induced defects (interstitial/vacancy groups) decrease and that they have completely disappeared by around 1750°C. Higher temperatures give the same result, the time taken to remove the defects being about one hour.

2.1.1 Rate of release curves

In most practical applications, it is the rate at which Wigner energy is released (dS/dt) that determines the temperature rise observed in a block of graphite and not the total stored energy, although this provides an upper limit to the energy available. When a dS/dt curve is measured as a function of annealing temperature in a linear rise calorimeter (see subsection 2.2), the temperature (T) is directly proportional to time. It is therefore possible to convert the dS/dt curve from a linear rise calorimeter into a dS/dT curve, which can then be compared with the specific heat of graphite. Figure 2.5 shows examples of dS/dT curves for graphite irradiated at 30°C to various fast neutron doses. These curves illustrate the formation of the 200°C peak, which at high doses broadens and reduces in height, although the total stored energy (represented by the area under the curve) continues to increase. Curves of dS/dT at an irradiation temperature of 150°C are shown in Figure 2.6 for various doses and Figure 2.7 shows typical dS/dT curves for different irradiation temperatures. The position of the 200°C dS/dT peak is also a function of the heating rate because of the different relaxation times associated with the interstitial complexes that make up the peak. This is shown in Figure 2.8. Overall, Wigner energy release occurs more slowly at lower rates of temperature rise.

Annealing of graphite irradiated above ~180°C (the lowest steady irradiation temperature in a Magnox reactor) does not show the 200°C peak but instead shows a "plateau" below the high temperature peak (see Figure 2.4) associated with annealing the larger interstitial complexes.

2.1.2 Saturation

As can be seen in Figure 2.3, the total stored energy appears to saturate at high doses (at least for the 30°C and 155°C curves). In parts of the older Magnox stations the stored energy may have saturated.

2.1.3 Radiation annealing

Early experiments in the USA suggested that irradiated graphite anneals to a greater extent when held at 225°C in a reactor than when held at the same temperature outside a reactor. According to Nightingale (1962) several samples of graphite were irradiated to various doses at 30°C, then annealed at 375° for several days and then re-irradiated at 335°C. The effect of this treatment on the total stored energy is shown in Figure 2.9. It is clear that the effect of annealing in the reactor (radiation annealing) is quite different from thermal annealing alone. Recent Russian work (Nikolaenko and Karpukhin 1996) shows a similar effect, and attributes it to the exposure to gamma radiation. On balance, it seems unlikely that this would be a significant effect under repository conditions, as discussed in subsection 2.7.

2.1.4 Effect of thermal annealing

Bridge *et al.* (1962) give the results of linear rise measurements using samples of graphite containing Wigner energy with a range of activation energies. Their experiments used several samples of graphite which had experienced the same irradiation treatment. One of these samples was measured as received after irradiation and the other samples were measured after heating for various times at constant temperature. The results of the measurements are shown in Figure 2.10. Bridge *et al.* calculated the spectrum using the results from the untreated specimen. They then calculated the activation energy spectrum to be expected after each heat treatment and from the spectrum they predicted the linear rise curves. Their results agree reasonably well with the observations, showing that the variable activation energy model (Vand 1943) can be used to predict complicated annealing behaviour, provided that the variation of temperature with time is known. These data could be considered for future validation studies on models of Wigner energy release.

2.2 Experimental Assay of Wigner Energy

There are many methods of measuring Wigner energy in irradiated graphite, but this review has deliberately limited itself to the bomb calorimeter, the adiabatic calorimeter (or dipping method) and the differential scanning calorimeter, since collectively these account for the majority of data available.

An empirical linear relationship has been found between total stored energy and the fractional change in thermal conductivity that occurs when graphite is irradiated. The measurement of thermal conductivity is quick and relatively easy and could be used to provide a rough estimate of the total stored energy in a piece of graphite (see below).

The choice of measurement method depends on the information required. For most disposal applications the rate of release of Wigner energy as a function of temperature at various heating rates will be the most relevant, since this determines the rate at which heat is released into the waste package and the temperature reached in the package is then determined by the rate at which heat is lost from the package. However, the total stored energy (S), which is the integral of the rate of release, can also be a useful parameter, since the total stored energy provides an upper limit to the energy available. For example, the specific heat capacity of graphite at 20°C is approximately 0.7 J $g^{-1}K^{-1}$, so if a block of graphite has a total stored energy of 8 J g^{-1} then the maximum temperature rise caused by the release of all the energy in the specimen would be 8/0.7 = 11.4 K. The specific heat of graphite increases with temperature, so at a higher temperature, a release of 8 J g^{-1} would produce a smaller temperature rise.

It may be worth noting that the maximum total stored energy measured in graphite from the Windscale Piles is about 1000 J g^{-1} (Marsden *et al*. 1997), which is small compared to the amount of heat that could be released by burning the graphite, 32,657 J g^{-1} (Nightingale 1962).

The measurement methods are briefly described in the following section; more comprehensive descriptions of some of the apparatus can be found in Simmons (1965).

2.2.1 The bomb calorimeter

The bomb calorimeter is the standard method of determining the calorific value of a combustible material. It consists simply of a small pressure vessel (the 'bomb'), into which is placed a known mass (usually 1 or 2 grams) of the substance to be tested. The bomb is then filled with an atmosphere of pure oxygen, and placed in a well insulated, stirred water bath, the temperature of which is measured using a platinum resistance thermometer. The system is allowed to reach thermal equilibrium before the content of the bomb is ignited by means of a small electrical spark plug built into the bomb. Again the system is allowed to reach thermal equilibrium and the difference in temperature before and after the bomb is fired is related to the amount of heat generated by the combustion of the material in the bomb. The system is usually calibrated by burning a known amount of benzoic acid in the bomb. Corrections are necessary for heat losses from the calorimeter and any unburnt material left in the bomb.

If graphite containing Wigner energy is burnt in a bomb calorimeter an excess amount of energy is liberated, above that which would have been produced by burning an equal amount of unirradiated graphite. This difference is the total amount of stored energy (S) in the graphite.

One disadvantage of the bomb calorimeter is that it only measures the total amount of energy released (in J g^{-1}), it does not provide any information about the temperature at which the energy is released. So for example, a bomb calorimeter could determine that a particular irradiated graphite specimen contained 100 J g^{-1} of Wigner energy, but it could not differentiate between the 200°C peak and the 1200°C peak. However, by preparing a series of specimens from the same piece of graphite (assuming that they all contain the same amount of Wigner energy and have the same release characteristics) and annealing these at different fixed temperatures for long periods of time (to ensure that all the energy releasable at that temperature had been released), it is possible to measure the amount of energy remaining in the specimens using a bomb calorimeter, thus obtaining a crude energy versus temperature distribution (Figure 2.11).

However, because two large numbers are being subtracted (heat of combustion of graphite with stored energy – heat of combustion of graphite without stored energy), the uncertainty on the measurement is significant at around ±12 (σ) J g^{-1} (Bridge *et al.* 1962).

Advantages

The bomb calorimeter is a standard piece of commercial equipment found in many laboratories.

Disadvantages

The bomb calorimeter only measures S (in J g^{-1}). It takes a long time for the calorimeter to reach thermal equilibrium, so typically only one or two specimens can be measured per day. There is quite a large uncertainty on the measurement.

2.2.2 The adiabatic calorimeter (or 'dipping' method)

In its crudest form, the method consists of attaching a specimen of irradiated graphite to a thermocouple and then 'dipping' the specimen into an oven at a constant temperature and monitoring the output from the thermocouple to see if the temperature of the graphite increases due to a release of Wigner energy. The specimen could be 'dipped' into a series of ovens at progressively higher temperatures until a release was observed (Figure 2.12). This method was popular in the early 50's as it provided the most realistic approximation to the behaviour of the graphite in a reactor during a Wigner release.

More elaborate apparatus has been described by Henson and Simmons (1959) and Rappeneau *et al*. (1962) with guard heaters that follow the temperature of the specimen to minimise heat losses, but they lack the simplicity of the 'dipping' technique.

Advantages

A quick and simple approximation to the behaviour of graphite in a reactor containing Wigner energy.

Disadvantages

The experimental boundary conditions are not well defined in the simple 'dipping' technique. The rate at which the temperature increases is determined by the size of the specimens and heat losses into the oven. The more complex apparatus provides better control of the boundary conditions, but use of the method was made largely redundant by the introduction of the linear rise calorimeter.

2.2.3 The differential scanning calorimeter

The vast majority of stored energy measurements made since the late 50's have been made with the temperature of the specimens rising at an approximately constant rate. Several types of apparatus have been devised for this measurement, starting with the early Sykes calorimeter (Sykes 1935) improved by Neubert and Lees in 1957, through to the controlled linear rise calorimeter described by Cottrell *et al.* in 1958 and by Henson and Mounsey in 1961. The modern equivalent of these linear rise calorimeters is the differential scanning calorimeter (DSC) manufactured by Perkin-Elmer and this is the instrument that will be described here. It should be noted that many manufacturers produce DSCs, but only the Perkin-Elmer model uses the power compensating controlled linear rise principle.

The Perkin-Elmer DSC consists of a water cooled aluminium block (maintained at a constant temperature) containing two platinum crucibles. The base of each platinum crucible contains two platinum resistance spirals, one that acts as a platinum resistance thermometer and one that acts as a platinum resistance heater (Figure 2.13a). The electronics in the calorimeter continually monitor the temperature of the two crucibles and adjust the power inputs to the heaters to keep them at the same temperature. The output from the calorimeter consists of the difference in power supplied to the two platinum crucibles and the temperature of the crucibles. A computer allows the temperature of the crucibles to be ramped between two fixed temperatures at predefined heating rates in the range 0.1 and 300 K min^{-1} (most measurements are made at 10 K min^{-1} because this allows them to be completed within a day) and processes the results. (In contrast, the ramp in temperature experienced by waste in a repository would be below this range, with a maximum of about 5×10^{-3} K min^{-1} (Guppy *et al*. 1999).)

The Perkin-Elmer DSC can be used to measure the rate of release of Wigner energy in various ways. The most basic method requires three separate measurement runs (each run must be over the same temperature range and at the same heating rate, see Figure 2.13b). The first run is made with the crucibles empty and is referred to as the "baseline". The second run is made with a known mass of reference material with a well-known specific heat (usually synthetic white sapphire) which is referred to as the "standard" and the third run is made with the reference material replaced by a known mass of specimen (in this case irradiated graphite) referred to as the "sample". The ratio in differential power between the "standard – baseline" and the "sample- baseline" is the same as the ratio of the specific heat of the reference material to that of the specimen, at any given temperature. In this way the specific heat of the graphite can be measured directly. If Wigner energy is released by the graphite, this shows up as a reduction in the specific heat of the material and the true specific heat can be obtained by performing a fourth measurement run, under identical conditions to the sample run, only this time any Wigner energy that can be annealed will already have been released.

An improved method of measuring the rate of Wigner energy release in a Perkin-Elmer DSC involves placing a similar mass of unirradiated graphite in one of the crucibles when the specimen is being run and then performing a fourth measurement run as described above. This eliminates the subtraction of the two large specific heat values obtained in the previous method to obtain the Wigner energy release rates.

Perkin-Elmer claim a sensitivity of ±8 μW for their DSC which for a typical specimen mass of 0.1 grams gives a maximum sensitivity of 80 $\mu J\ g^{-1}\ s^{-1}$, however this does not make any allowance for drift of the apparatus with time. Allowing for measurement being made over the course of a day (40 to 600°C at a heating rate of 10 $K\ min^{-1}$) a sensitivity of 0.001 $J\ g^{-1}\ s^{-1}$ is considered more achievable.

Advantages

The Perkin-Elmer DSC is commercially available and is capable of operating from –173°C to +723°C. It measures rate of release directly, with no time lag. It only requires a small amount of sample, typically ~0.1 gram.

Disadvantages

The apparatus drifts due to ambient temperature variations and with time. It is assumed that no energy is being released at the start and end of the scan (it can be programmed to hold at these temperatures to allow energy to be released before starting a run). The low end of the rate of heating of samples achievable in the laboratory is appreciably above the heating rate that would exist in a repository.

2.2.4 Thermal conductivity

It should be noted that at low doses (Bell et al. 1962) there is a linear relation (Figure 2.14) between total stored energy and the fractional change in thermal conductivity

$$S = 6.25\left(\frac{K_{unirradiated}}{K_{irradiated}} - 1\right)$$

where S is the total stored energy in Cal. g^{-1}
$K_{unirradiated}$ is the thermal conductivity of unirradiated graphite
and $K_{irradiated}$ is the thermal conductivity of irradiated graphite

Consequently, measuring the thermal conductivity of the graphite can provide a crude estimate of the total stored energy in the graphite. Thermal conductivity can be measured quickly using the laser flash technique (BSI 7134:Part 4.2:1990) and provides a rapid means of assessing the approximate total stored energy in a piece of irradiated graphite provided the unirradiated thermal conductivity is known. However, at high doses total stored energy saturates (Figure 2.3) and this relation may breakdown. Similarly, this relation may not hold for graphite removed from the Windscale Piles that have experienced several anneals during their operation.

2.3 Limitations of Assay Techniques

These are described for the three main techniques in Table 2.1, which also summarises the other main features of the methods.

2.4 Assessment of Wigner Energy Release by Modelling

Following the discovery of Wigner energy in graphite and especially after the Windscale fire, considerable effort was devoted to studying Wigner energy and trying to model (and hence predict) its behaviour. Being a thermally activated process, a natural starting point was the Boltzmann distribution:

$$N = N_0 \exp\left(-\frac{E}{kT}\right)$$

which describes the number of atoms or molecules in a substance with a thermal energy E above the ground level, in terms of the number of molecules at ground level N_0, Boltmann's constant k and the absolute temperature T. This relation is also known as the Arrhenius equation and is widely used to describe the rate of a chemical reaction as a function of temperature. In chemical kinetics, E represents the activation energy for the reaction.

The Boltzmann function (above) can be applied to the annealing of Wigner energy in several ways, depending on whether the activation energy *E*, the pre-exponential term or both are varied. The various options are discussed by Simmons (1965) and are summarised below.

a) <u>Constant activation energy</u>

The basic Boltzmann equation (as described above) has been widely used (Cottrell *et al.* 1958) to illustrate the effect of various parameters on stored energy release with an activation energy *E*=1.7 eV. It also lends itself to the rapid determination of the activation energy *E*, since if two identical samples are maintained at constant temperatures T_1 and T_2 then:

$$\left(\frac{dS}{dt}\right)_1 = F(S)\exp\left(-\frac{E}{kT_1}\right)$$

$$\left(\frac{dS}{dt}\right)_2 = F(S)\exp\left(-\frac{E}{kT_2}\right)$$

and when *S* has the same value in both samples:

$$\ln\left[\frac{\left(dS/dt\right)_1}{\left(dS/dt\right)_2}\right] = \frac{E}{k}\left[\frac{1}{T_2} - \frac{1}{T_1}\right]$$

A measurement of $\left(\frac{dS}{dt}\right)_a$ at two very different heating rates a_1 and a_2 also yields the activation energy *E*. Determining the rates and temperatures at which *S* is the same, then:

$$\left(\frac{dS}{dT}\right)_1 = \frac{F(S)}{a_1}\exp\left(-\frac{E}{kT_1}\right)$$

$$\left(\frac{dS}{dT}\right)_2 = \frac{F(S)}{a_2}\exp\left(-\frac{E}{kT_2}\right)$$

or

$$\frac{E}{k}\left[\frac{1}{T_2} - \frac{1}{T_1}\right] = \ln\left[\frac{a_1\left(dS/dT\right)_1}{a_2\left(dS/dT\right)_2}\right]$$

Because of the logarithmic term in the above equation a large difference in heating rates is required to determine E accurately.

Another variation of the constant activation energy approach used in the USA is to apply an equation of the form:

$$\frac{dS}{dt} = -A\,S^{\gamma}\exp\left(-\frac{E}{k\,T}\right)$$

where S is the stored energy and γ ranges between 6 and 8.

However, although a single activation energy can be used to approximate the form of a rate of release peak, it cannot produce the broad peaks or the continuum observed in rate of release measurements on reactor irradiated graphite.

b) Variable activation energy / constant frequency factor

This model, due originally to Vand (1943) and developed by Primak (1955, 1956) assumes in its simplest form that the energy release process for each group of defects can be described by an equation of the form

$$\frac{dS}{dt}(E,t) = -\nu\,S(E,t)\exp\left(-\frac{E}{k\,T}\right)$$

where ν is a constant frequency factor, *S* is the stored energy with activation energy *E*, at time *t*.

Using this model Bridge *et al*. (1962) derived a thermal relaxation time τ (effectively the lifetime) for defects with different activation energies:

$$\tau = \frac{1}{A\exp\left(-\frac{E}{kT}\right)}$$

Bridge used these models to predict the effect of thermal annealing on $\frac{dS}{dt}$ and more recently Minshall (1999) has used them to predict the $\frac{dS}{dt}$ curves at different heating rates (see below).

c) Variable frequency factor/Constant activation energy

In this case it is assumed that all the stored energy is released according to a sum of first order processes with constant activation energy E, but variable frequency factors, i.e. each process is

$$\frac{dS}{dt}(\ln \nu) = -S(\ln \nu)\ \nu \exp\left(-\frac{E}{kT}\right)$$

The total stored energy *S* is the sum of all the contributions with different values of ν. Very little work has been done on this type of model.

Minshall (1999) notes that the concept of a "start temperature" below which no stored Wigner energy is released is incorrect. Based upon first order kinetics, which are believed to represent the process reasonably well, there is no temperature below which no release can occur, rather the release is so small as to not be detectable. The paper also points out that to consider the extent of any release, the heat transfer to the environment of the graphite must be accounted for. These factors should be taken into account in modelling the release of Wigner energy in the environment of a repository, and the implications this has for temperature rises.

The paper goes on to review the classic papers on the theory of stored Wigner energy release and concludes that to achieve better accuracy than hitherto, a rate of release model must account for the spread of activation energies within any graphite sample. The following expression for the rate of release of energy which describes a multi-activation energy release model is therefore proposed:

$$\frac{dS}{dt} = -\nu \sum_i S(E_i) \exp\left(\frac{-E_i}{kT}\right)$$

where

$\frac{dS}{dt}$ is the rate of release of stored energy $S(E_i)$

E_i is the activation energy associated with the stored energy $S(E_i)$

k, ν, T are Boltzmann's constant, a frequency factor and absolute temperature respectively

The Minshall model is the most suitable available for assessing Wigner energy release from irradiated graphite. Guppy *et al.* (1999) have used it to calculate the release of Wigner energy within a finite element model of the Nirex Deep Waste Repository vault concept. This work is discussed in more detail in subsection 2.7.

Wörner *et al.* (2000) report on the use of Minshall's multi-activation energy model to estimate the sensitivity of Windscale Pile 1 graphite to stored energy release whilst being grouted for disposal. The conclusion is reached that some of the graphite may start to release significant amounts of energy whilst being grouted.

Botzem *et al.* (1999) report on work that calculated the temperatures that may occur in boxes of grouted graphite within a Nirex repository. The results show that it is very difficult to show that high temperature excursions will not occur and that it is unclear whether graphite containing Wigner energy is consistent with the regulatory views on ‘passive safety’.

2.5 Options for Managing Graphite Wastes

Four main routes are discussed in the literature for managing irradiated graphite wastes:

- Direct disposal of the graphite in containers to a repository; the waste may be either grouted into the disposal container or ungrouted.
- Incineration, with disposal of the small volume of residue.
- Steam reforming (a form of pyrolysis), which like incineration converts the bulk of the graphite to gas and leaves a residue for disposal.
- Annealing out the Wigner energy at around 200°C by heating, followed by packaging and disposal to a repository.

Although the graphite might in principle be recycled (as mentioned briefly by Neighbour *et al*. 2000), the associated processing before reuse could be costly given that the material is radioactive and contains Wigner energy. In addition, its physical properties will have been altered during irradiation and may not be appropriate to potential applications.

All the routes except direct disposal would generate secondary wastes that could either be discharged to the environment (notably gases containing tritium and carbon-14 (Wise 2000)) or, with appropriate off-gas treatment, converted to a material that could be sent for disposal.

The papers identified consider in some detail how these processes could be implemented and to some extent how they compare. None, however, contains a full comparison of all the options. In addition, waste producers should also consider the 'do nothing' option, as recommended by the Environment Agency (2000): this is discussed in Section 6.

Wise (2000) discusses the irradiated graphite liabilities arising from the United Kingdom Atomic Energy Authority’s (UKAEA’s) nuclear programme and likely management routes. Of the reactors currently planned for decommissioning, most have substantial graphite arisings, although not all raise concerns as regards Wigner energy.

	Mass of Graphite (tonnes)	Wigner Energy Considered to be an Issue	Currently Planned Disposal route
GLEEP	505	No	Disposal as LLW in half height ISO containers at Drigg
BEPO	766	Yes	Not defined. Incineration and disposal as ILW at Nirex repository under consideration
Windscale Pile 1	1966	Yes	Disposal as ILW at Nirex repository after annealing. Large blocks to be annealed. Undamaged graphite in 4m boxes. Damaged graphite in $3m^3$ boxes.
Windscale Pile 2	1966	Yes	No current plans, although likely that any plan would follow that for Pile 1. Continued safe storage planned.
Dounreay Fast Reactor	200	No	Disposal as ILW at Nirex repository in 4m boxes.
Prototype Fast Reactor	To be assessed	No	Disposal as ILW at Nirex repository in 4m boxes.
Material Test Reactors	17	Yes	Disposal as ILW at Nirex repository in 4m boxes.
Windscale Advanced Gas Cooled Reactor	210	No	Disposal as ILW at Nirex repository in WAGR boxes

As shown above the options considered are incineration (followed by disposal of the small volume of residue) or direct disposal either as LLW at Drigg or ILW to a deep repository. Three methods of incineration are discussed (conventional burning, fluidised bed, power laser driven) but it is concluded that incineration is not favoured because of the impact upon the environment from radiological and non-radiological gaseous emissions, the generation of secondary wastes and the public perception issues. Disposal as either LLW or ILW waste is therefore the preferred option. Only graphite waste from GLEEP is considered suitable for disposal at Drigg, due to its low specific activity. For the graphite wastes from the other reactors, each reactor is being separately assessed to consider whether the Wigner energy will require annealing prior to disposal in the Nirex repository. This requires sampling and measurement of the Wigner energy content of the graphite and then assessment by Nirex using the Nirex thermal vault models (Guppy *et al.* 1999).

Wise (2000) also discusses briefly the immobilisation of the graphite dust that will be generated during decommissioning of graphite structures. It is concluded that this area is at an early stage of development. A wetting agent would be required to achieve adequate wetting of the dust by a cement grout. It has been found that small amounts of dust can be incorporated into a cement matrix. Other options under consideration include supercompaction of the dust, use of a polymer encapsulant, and containment of the dust before cementation.

Wickham *et al*. (1998) state that the UK's Advanced Gas Cooled Reactors (AGRs) are operated at too high a temperature for there to be a significant accumulation of Wigner energy. Some stored energy does accumulate in the lower part of Magnox reactor cores, but the total is insufficient to lead to self-heating in any circumstances. It is claimed that there is no possible energy release hazard from frictional heating or other means whilst handling.

Neighbour *et al.* (2000) briefly consider the worldwide inventory of irradiated graphite and the recent re-emergence of designs for a High Temperature Gas Cooled reactors with graphite reflectors (in which design Wigner energy is not considered to be an issue because of the high temperature of operation and hence self-annealing behaviour). A full list of options for the disposal of graphite is listed and discussed. The specific issues surrounding Wigner energy in any of the graphite wastes are not addressed however. A discussion is provided of the assessment of the environmental impact of the release of ^{14}C and (with incineration) carbon dioxide to the environment in managing graphite wastes and how this might be modified by the perception of the issue by the public.

Holt (1999) describes the decommissioning strategy for Magnox reactors. Incineration and encapsulation are the two main options for treating the graphite waste stream that has arisen from graphite fuel debris at Berkeley and Hunterston. It is planned to follow the Safestore strategy for the reactor cores, with decommissioning taking place 135 years after reactor shut-down. It is stated that Wigner energy has been considered as part of the planning, but no details are given. Incineration and encapsulation are also considered for the eventual treatment of the cores. It is noted that assessment studies appear to preclude the discharge of radioactive ^{14}C as a gas generated by incineration and hence encapsulation is the preferred technique. Disposal in a deep geological repository is the preferred eventual disposal route, preferred over shallow land burial because of the risk of impact of ^{14}C release (Department of the Environment 1986).

Marsden *et al.* (1997) present the results of surveys of the graphite core of Windscale Piles 1 and 2. Visual surveys were carried out inside the core by traversing specially designed cameras along the fuel channels. Samples were then trepanned from selected locations and measurements of density, activity, thermal conductivity and stored Wigner energy made. The paper discusses the issues behind the trepanning of samples from the cores, in particular the possibility of initiating a significant release of Wigner energy or an explosion of the graphite dust. It is concluded that the risks of either are small. From Pile 2 results, levels of total stored Wigner energy up to 1000 J g^{-1} were measured by bomb calorimetry. Using differential scanning calorimetry with an increase in temperature of 10 K min^{-1}, the graphite specimens showed a peak in stored energy release rate at around 200°C with no measurable release below 88°C. When written, the sampling of Pile 1 was incomplete. No samples were planned to be taken within the zone damaged by the fire in Pile 1.

Wörner *et al.* (2000) also present results of the stored energy survey of Windscale Pile 1. They report that whereas some graphite shows energy release curves that exceed the specific heat of the unirradiated graphite a lot of the graphite does not. They also note that the stored energy in some graphite is still unknown because samples could not be taken.

The following statistical evaluation of the survey data is presented:

- 11 samples (23%) are characterised by the '200°C' peak. In 6% of the samples this peak is shifted into the temperature range 250°C to 300°C.
- 30 samples (64%) are characterised by a flat peak between 300°C and 400°C.
- 6 samples (13%) are characterised by an energy release which begins to be measurable above 400°C.

The authors conclude that 77% of the investigated material does not represent a risk because neither handling nor encapsulation operations will result in a Wigner energy release, given the temperatures involved. However because the graphite that does pose a risk of Wigner energy release cannot be identified except with an impractically large amount of analysis, it will be necessary to anneal all the larger pieces (Wörner *et al,* 2000). The authors then describe investigations to determine the parameters for an annealing process.

The investigations were conducted on a dowel from Pile 2 and graphite samples from the dowel were heated at 25K min^{-1} from 40°C to successively higher temperatures up to 400°C. The maximum temperature was held for three hours, after which the sample was cooled, left in liquid nitrogen for one week and then tested again to 100K above the previous test temperature. Results obtained showed that by 400°C most of the energy in the sample had been released by the first test and that no significant amount of energy was released in any of the second tests until the temperature was 50K above the previous test temperature. In terms of total stored energy it was found that annealing to 200°C releases 55% of the stored energy, to 250°C releases 78% and to 300°C releases 90%. It is concluded that heating the graphite to between 250°C and 300°C for 30 minutes will remove between 80% and 90% of the stored energy releasable up to 500°C and will provide adequate process conditions for annealing Windscale Pile 1 graphite.

Botzem *et al.* (1999) describe work that forms part of the Pile 1 decommissioning project to develop an annealing plant as part of the waste processing facility. The plant design draws on the work by Wörner *et al.* (2000) to determine the process parameters for annealing. The annealing process would occur in an inert argon atmosphere and only undamaged graphite, i.e. undamaged from the fire, would be annealed. The six annealing ovens would process a full size core block (210 mm x 210 mm x 790 mm) or two half size blocks every ten minutes. The ovens would operate in parallel: this would ensure the desired throughput of graphite. The graphite would be heated to a maximum of 250ºC and then cooled to less than 50°C in an air cooling station. The extent of graphite oxidation by chemical contaminants would help to determine the appropriateness of this final step. After annealing the waste would be packaged into standard Nirex 4 m ILW boxes with an estimated loading of 8.3 to 8.7 m^3 per box.

The process is designed to remove the Wigner energy associated with a release temperature of around 200ºC, the fraction of the total stored energy that might be released under repository conditions (see Section 5).

A study of possible heating methods rejected convective heating because it is too slow and radiative heating because of the need to replace the radiation heaters during the lifetime of the plant and the low thermal conductivity of the irradiated graphite results in poor penetration of heat to the centre of the blocks. The preferred heating method is induction heating which generates heat directly within the graphite. Tests on unirradiated graphite samples of representative size showed temperatures of 350°C were achieved within 15 minutes with a power input of 30 kW at a frequency of 3 kHz. These conditions will require optimisation using irradiated graphite because of its lower thermal conductivity.

It is proposed to heat the graphite in the ovens to surface temperature of 200°C within approximately 7 minutes, pause for 3 minutes and then raise the temperature to 250°C. This temperature would be maintained for 20 minutes to observe behaviour. If Wigner energy raises the temperature the heating would be switched off. The proposed heating regime has been assessed from experimental work on annealing Wigner energy from samples of the Pile 1 graphite.

Release of tritium from the graphite during annealing has been examined experimentally by Wörner et al. (2000). It was concluded that under the anticipated operating conditions, less than 0.5% of the tritium would be released from the plant. In the experiments, the gas was released as both tritiated water and hydrogen. It appears that this would be discharged to the atmosphere, and the case for doing this still needs to be made in terms of system optimisation. Tritium, ^{14}C and ^{137}Cs dominate the radionuclide inventory of the Pile 1 graphite (Wise 2000). No data are presented for the release of gases containing ^{14}C and ^{137}Cs or for particulate releases.

In terms of safety during final disposal, it may be possible to fill containers with annealed graphite (i.e. without grout). The level of residual Wigner energy may be low enough to prevent a significant temperature rise in the disposal vaults. This assumes that there would be no redistribution ('trickle down') of high-temperature Wigner energy to levels with lower activation energies governing release (see subsections 2.1.3 and 2.7).

Mason *et al*. (2000) present pyrolysis as an alternative disposal route for graphite reactor cores. It is asserted that disposal of solid graphite presents 'interesting challenges' because of the long half-life of carbon-14 coupled with the innate potential of graphite to be converted to gaseous forms. Deliberate, early release of the gaseous carbon-14 (by incineration for example) may be acceptable on the basis that large amounts of the isotope occur naturally, but viewed as the total additional collective dose the proposition becomes unacceptable. It would also be essential to minimise the release of non-carbon isotopes such as tritium, iron-55 and cobalt-60. Disposal at sea provides better dilution of the carbon-14, but is currently unacceptable.

Pyrolysis of graphite involves steam heating of a graphite slurry in water in the presence of insufficient or no oxygen. The graphite is first powdered (unless it has been generated in the form of graphite dust) before conversion to a slurry. The gaseous products can then be oxidised to produce carbon dioxide and water. It is claimed that the process offers a number of benefits over combustion, such as tightly controlled containment, low volume of off-gas, separation of carbon from other radioactive elements, tight control over the process rate, and the possibility of applying to graphite *in situ*. Two methods of achieving this last benefit are presented.

The process operates at a few hundred °C, sufficient to release the low temperature Wigner energy. It is unclear how controllable the process would be under these conditions.

The paper suggests that reacting the carbon dioxide formed from incineration with calcium or magnesium oxide, hydroxide or metal to form a stable carbonate solid is a further possibility. A possible source of magnesium is the Magnox fuel cladding. Although this increases the volume of waste it is proposed that the solid could be used as a void filler in grouting or other radioactive waste. Tritium in the wastes could be converted to water.

Barlow *et al*. (1998) describe how Nirex is responsible for developing facilities for the safe disposal of intermediate and certain low level waste (including graphite) within the UK. Near to the commencement of operation of a repository, Nirex will issue waste acceptance criteria that all waste packages will have to comply with before being accepted for disposal. In the mean time Nirex is preparing specifications and guidance in order to permit wastes to be packaged in a form that is compatible with plans for transport and disposal.

Graphite wastes arise from the construction of reactor cores, in fuel elements and fuel stringers, as physical support for fuel elements (boats, as spacers and coolant flow modifiers (dowels)). For graphite wastes arising from most reactor systems the Wigner energy is not considered accessible until the graphite is heated to 'several hundred degrees centigrade'. Where the graphite was irradiated at temperatures close to ambient however (such as in prototype and experimental reactors), the stored energy can be released 'at relatively low temperatures'. A dose of greater than 1×10^{20} neutrons cm^{-2} is considered to result in a considerable amount of stored energy which can amount to 1 kJ g^{-1}.

2.6 Emplacement Strategies in a Repository

Little literature has been identified relating to emplacement strategies for individual types of waste in a deep repository for ILW. Barlow *et al.* (1998) note how a waste emplacement strategy may be necessary to control waste package temperature within a repository (and hence avoid the release of stored Wigner energy). Packages containing irradiated graphite might be positioned in such a way as to minimise the heat input to the packages and maximise the degree of control exerted over a release of the stored energy. With annealed graphite, an emplacement strategy could provide a second line of defence against release of stored energy.

The policy requirement to achieve passive safety in the management of waste packages is noted in subsection 1.1. It could be argued that relying on a particular pattern of emplacement in a repository that would be implemented some decades after the irradiated graphite has been packaged goes against this requirement, and that it would be preferable to manage the waste in such a way that repository safety could not be compromised by the location of the packages in the disposal vaults.

2.7 Release of Wigner Energy in a Repository

Marsden *et al.* (1995) briefly consider disposal of graphite containing Wigner energy and highlight that Wigner energy release may be a problem through raising the temperature of the waste, through the shrinkage of the graphite that occurs when the stored energy is released, and through the changes in other graphite properties (coefficient of thermal expansion, elastic modulus, strength and thermal conductivity). These factors could lead to physical and chemical changes in a repository, possibly leading to its safety being degraded.

Barlow *et al.* (1998) state that to prevent a rapid release of the stored Wigner Energy, it is necessary to ensure that the temperature of the graphite does not exceed an 'initiation' temperature. Control of package temperature after emplacement in a repository can be achieved by analysis of the radiolytic and chemical energy input to the repository, the repository design, ambient temperature, and a waste emplacement strategy. Temperatures of 30-35^{o}C arise in a host rock at a depth of around 500 m. Other heat sources are stated to yield a peak temperature of 50-100^{o}C. These temperatures are believed to be sufficient to release a 'significant fraction' of the stored energy in low temperature irradiated graphite.

Guppy *et al.* (1999) have implemented Minshall's variable activation energy release model for calculating the release of Wigner energy within a finite element model of the Nirex Deep Waste Repository vault concept. The modelling assumed that graphite waste was contained in Nirex 4 m boxes stacked in an array of three boxes wide and 6 boxes high occupying the whole vertical cross-section of a vault. Based on the assumption that the length of a vault would be much greater than its width, the heat flux along the vault was assumed to be negligibly small. Modelling therefore only needed to consider one vertical array of 18 packages. The graphite waste was assumed to come from the decommissioning of the Windscale Pile 1 and not to be annealed. The amount of low-temperature Wigner energy that it was assumed to contain constituted a worst case, based on experimental work on samples. Four cases are presented:

- Case A - a reference base case in which non-graphite wastes are represented by a typical cement encapsulant. The peak temperature reached in the waste package is approximately 58°C due to the heat generated from the curing of the backfilling material (a mix of Portland cement, calcium hydroxide, limestone flour and water), quoted as peaking at 2.7 kW m^{-3}. This occurs at the periphery of the waste after 4 days (i.e. where the package meets the backfill). The peak temperature at the centre of a waste package is 56°C occurring after 100 days and is associated with the conducted heat from the backfill, radiogenic decay and other heat sources.
- Case B - a box containing pure graphite waste was positioned near the centre of the waste stack (the most insulated location and hence the location that would be most affected by a release of Wigner energy). The peak temperature calculated in the waste exceeded 140°C when the calculation was stopped, the paper suggesting that the final temperature would have been appreciably higher because the temperature was continuing to rise rapidly and there was still a large fraction of the Wigner energy left in the graphite.
- Case C - the pure graphite waste in case B was replaced with mixture of 50% graphite and 50% inert cementitious material arranged in a chequerboard pattern. The peak temperature calculated in the waste was 78°C after 20 days.
- Case D - all boxes within the stack contain the mixture of 50% graphite and 50% inert cementitious material arranged in a chequerboard pattern. The peak temperature calculated in the waste was 95°C after 30 days.

It is concluded that, for the assumptions of design and properties made, 4 m boxes full of Pile graphite may behave in an unstable manner in the disposal environment. The dilution of graphite waste with an inert heat absorber however results in a more stable response.

Two cases considering a 1000°C fire lasting one hour and followed by a period of cooling in ambient conditions are also reported. In one case, the box contained graphite with stored energy and in the other it contained none. It is concluded that the Wigner energy increases the rate of temperature rise occurring in the graphite and suggested that all the graphite in the box would eventually release Wigner energy, although this was not explicitly demonstrated. The results are considered to be affected by the limited data available for the Wigner energy release model (only available up to 350°C).

The overall conclusion from the work (which is based in part on assumptions made concerning packaging arrangements and graphite properties) is that the potential for rapid release of Wigner energy from a waste package containing Windscale Pile graphite is not considered consistent with Nirex's requirements for inherent safety, passive immobilisation of radionuclides and robustness to uncertainties in the siting and design of a repository. This is particularly marked with the boxes containing 100% graphite, with no grout to act as a heat sink. Hence Nirex have recommended that Windscale Pile graphite should be annealed prior to packaging.

No other literature was identified on containers loaded with graphite and grout and the distribution of the packages. No assessments were identified relating to graphite that had been annealed before packaging.

It is noted in subsection 2.1.3 that annealing of graphite under reactor conditions may be influenced by the levels of γ-irradiation involved (Nikolaenko and Karpukhin 1996). This interpretation suggests that in a repository, γ-irradiation from the graphite wastes themselves and surrounding packages may lead to Wigner energy that previously could not be annealed being released. The γ-irradiation might cause energy levels that could subsequently be annealed at low temperatures to be populated. The subsequent energy release in a repository might then be higher than anticipated. If the graphite had previously been annealed at up to 200°C, a release might occur where none was expected.

A number of mitigating factors can be identified in the repository context:

- The dose rates in a repository would be very much lower than in a reactor.
- The container itself may provide some shielding, particularly if the irradiated graphite is disposed of in a shielded container.
- Any grout in the graphite package would provide some shielding.
- As would the backfill (but only after it had been emplaced) and grout in other packages.

Although on balance this appears to be an unimportant effect, it has yet to be addressed in the literature in a systematic way for its effect on the annealing and direct disposal routes.

2.8 Outstanding Issues

The following issues have been identified from the literature review as meriting further consideration in the comparison of options for managing irradiated graphite wastes, some of which are pursued later in the present report:

1. Publication in the open literature of work specific to the management of these wastes should continue. The BNES/IAEA conference in 1999 led to a considerable increase in the number of such publications, and although much has been achieved, further papers should appear as the subject matures to help build confidence in the management route(s) selected.
2. Prior removal by annealing of the Wigner energy that might otherwise be released in a repository would require a quality checking regime for the treated graphite to confirm that the process has performed as intended.
3. The effectiveness of annealing in terms of the temperature rise that any residual low temperature Wigner energy may cause has yet to be demonstrated, for example by modelling. This would form part of the justification for any proposal to package annealed graphite without grouting.
4. The release of radioactive gases containing tritium and possibly carbon-14 and particulates to the atmosphere from the annealing process proposed for the Pile 1 graphite requires further investigation and justification.
5. Graphite dust will be generated in decommissioning operations; work aimed at managing this waste is at an early stage.
6. Whilst on balance it appears unlikely that the γ-radiation fields in a repository could cause additional Wigner energy to be released under the prevailing conditions from directly-disposed and annealed graphite, confirmation is required of this.
7. No information exists on the comparative costs of the options identified for managing irradiated graphite. In order to compare options, this should include the total cost associated with all the various steps of each waste management system (e.g. so as to include the costs of disposal as well as any initial annealing and packaging).
8. A comprehensive study of the events in a repository that could lead to Wigner energy release appears not to have been reported. A initial study of this nature appears in Section 4 of this report.
9. The effects of Wigner energy release on post-closure safety (including how much can be tolerated) are not described in the literature. This issue is considered in Section 5 of this report.
10. An appreciable amount has been published concerning the implementation of the main options for managing irradiated graphite. However, a systematic basis for comparing options does not appear in the literature. Such a methodology will be required if preferred routes are to be demonstrated to represent Best Practicable Environmental Option. This is considered further in Section 6 of this paper.
11. The policy requirement to consider the practicability of reworking waste packages does not feature in the literature on managing irradiated graphite. This is considered in Section 6 of this paper.
12. A detailed assessment of the 'do nothing' option does not appear in the literature. This is considered further in Section 6.

Table 2.1 Advantages / disadvantages of the main assay techniques

	Bomb Calorimeter	**Adiabatic Calorimeter**	**Linear Rise Calorimeter**
Commercially available	Yes	Yes	Yes
Parameter determined	Total stored energy	Temperature rise	dS/dt or dS/dT
Duration of test	4 hours	2 hours	8 hours
Mass of sample (g)	1	10→1000	0.1
Accuracy	±12 $J.g^{-1}$	±1 K	0.001 $J.g^{-1}.sec^{-1}$

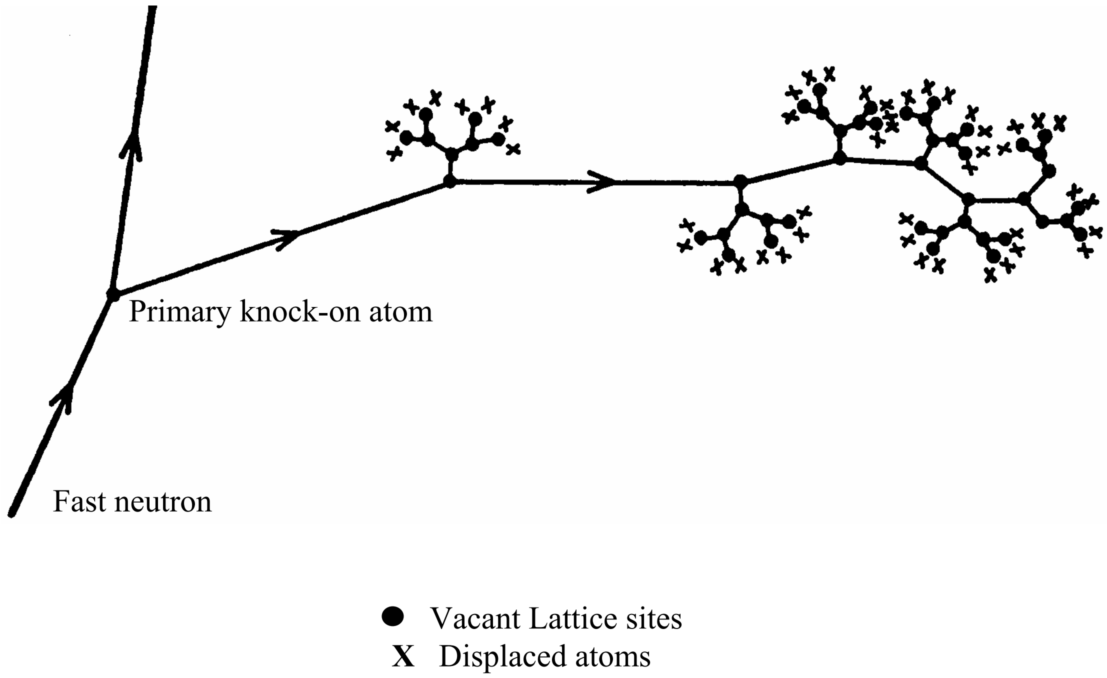
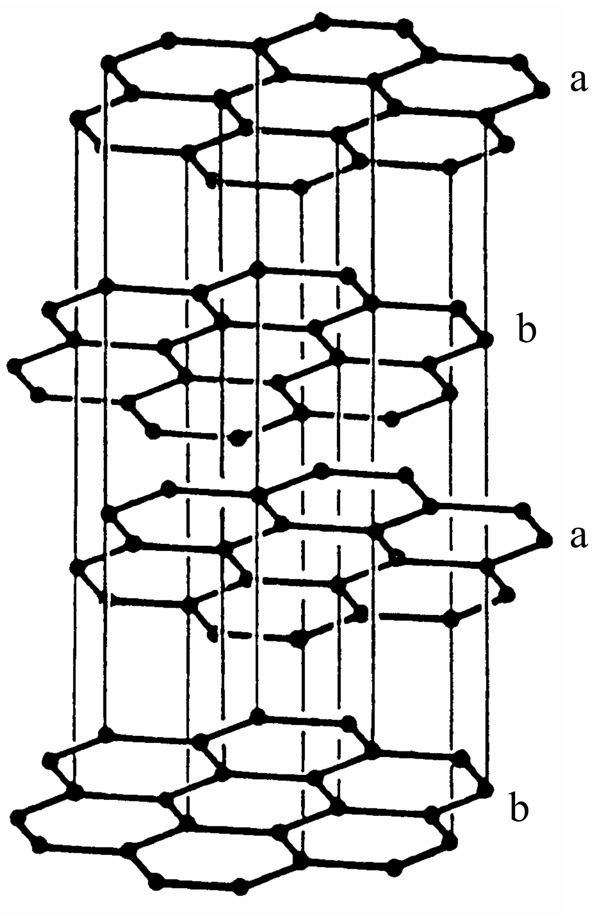


Figure 2.1 Distribution of displaced atoms and vacant lattice sites (from Simmons 1965)



The hexagonal structure of graphite

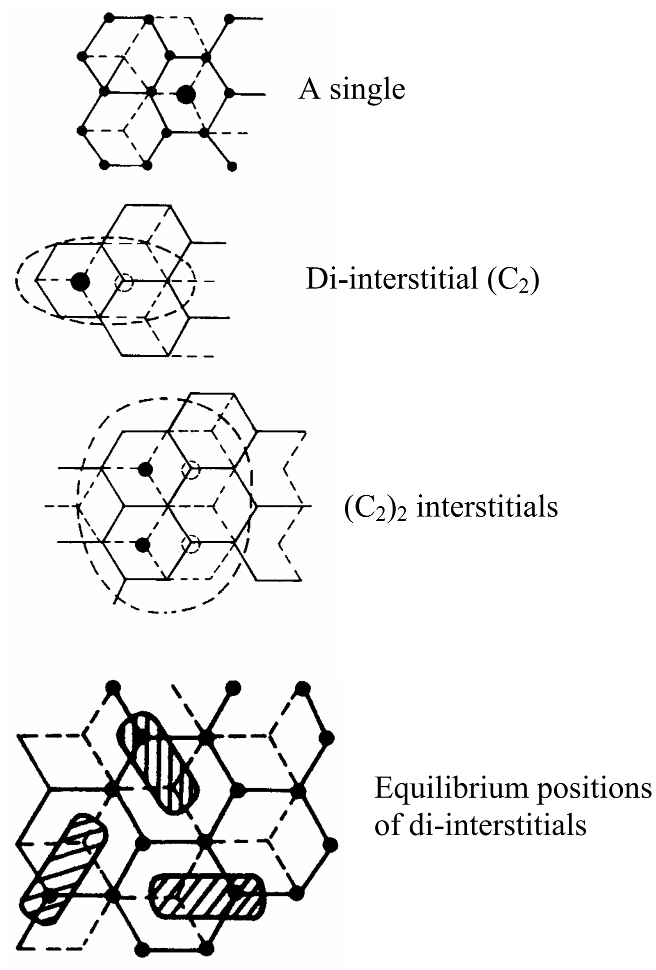


Figure 2.2 The structure of graphite and the formation of interstitials (from Simmons 1965 and Kelly 1981)

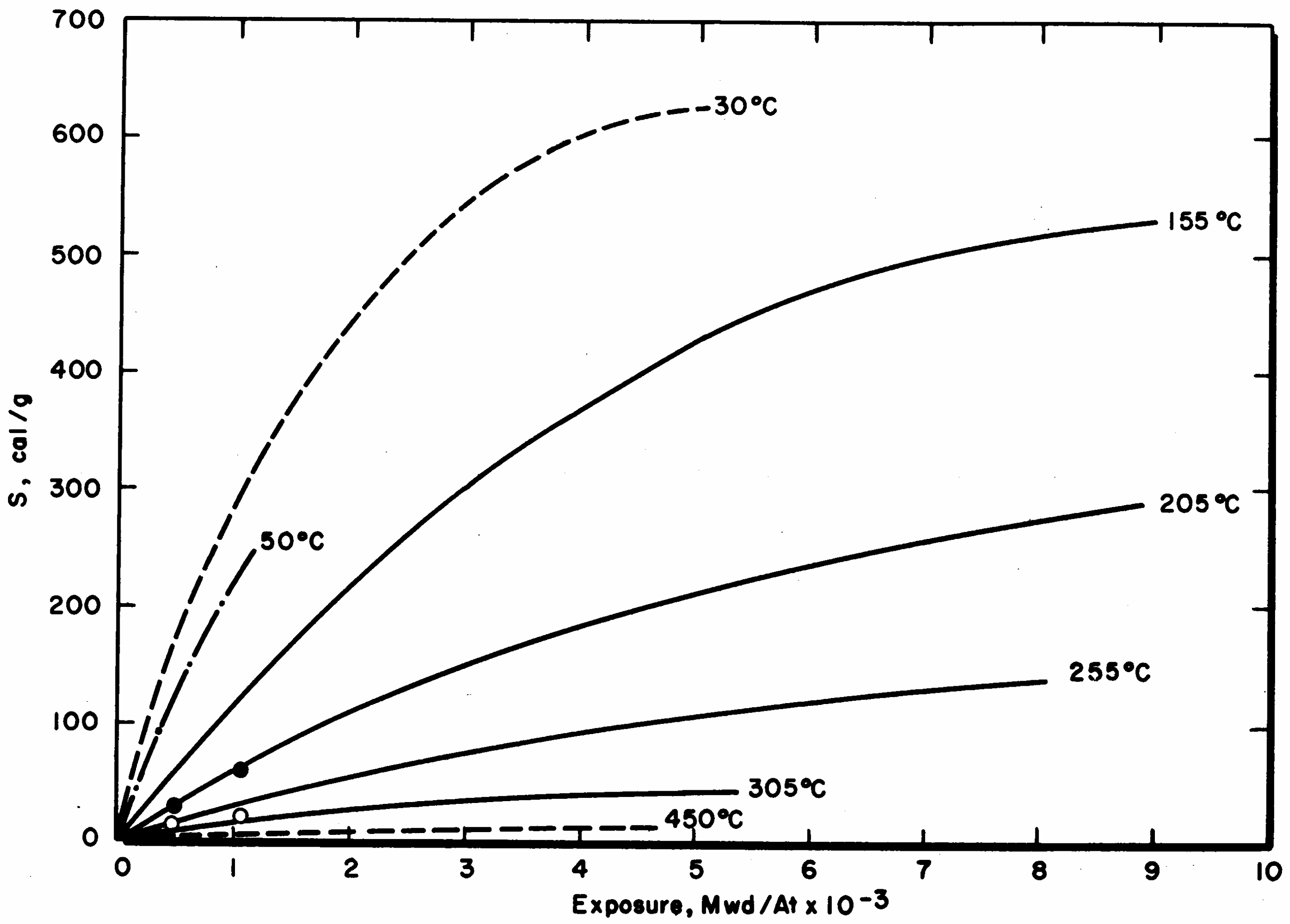


Figure 2.3 The accumulation of total stored energy (S) at various irradiation temperatures (from Nightingale 1962)

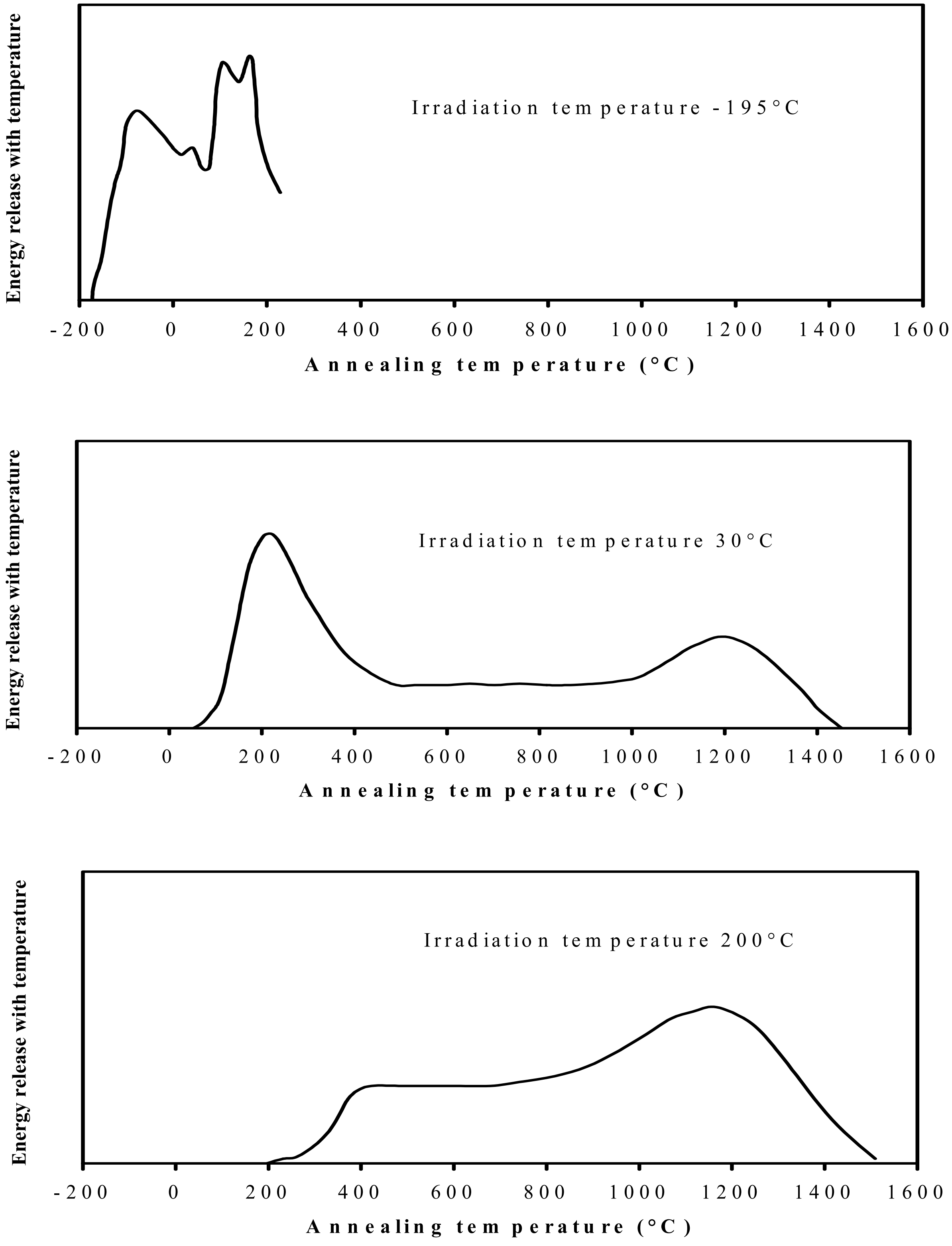


Figure 2.4 Schematic annealing spectra for graphite irradiated at various temperatures (from Simmons 1965)

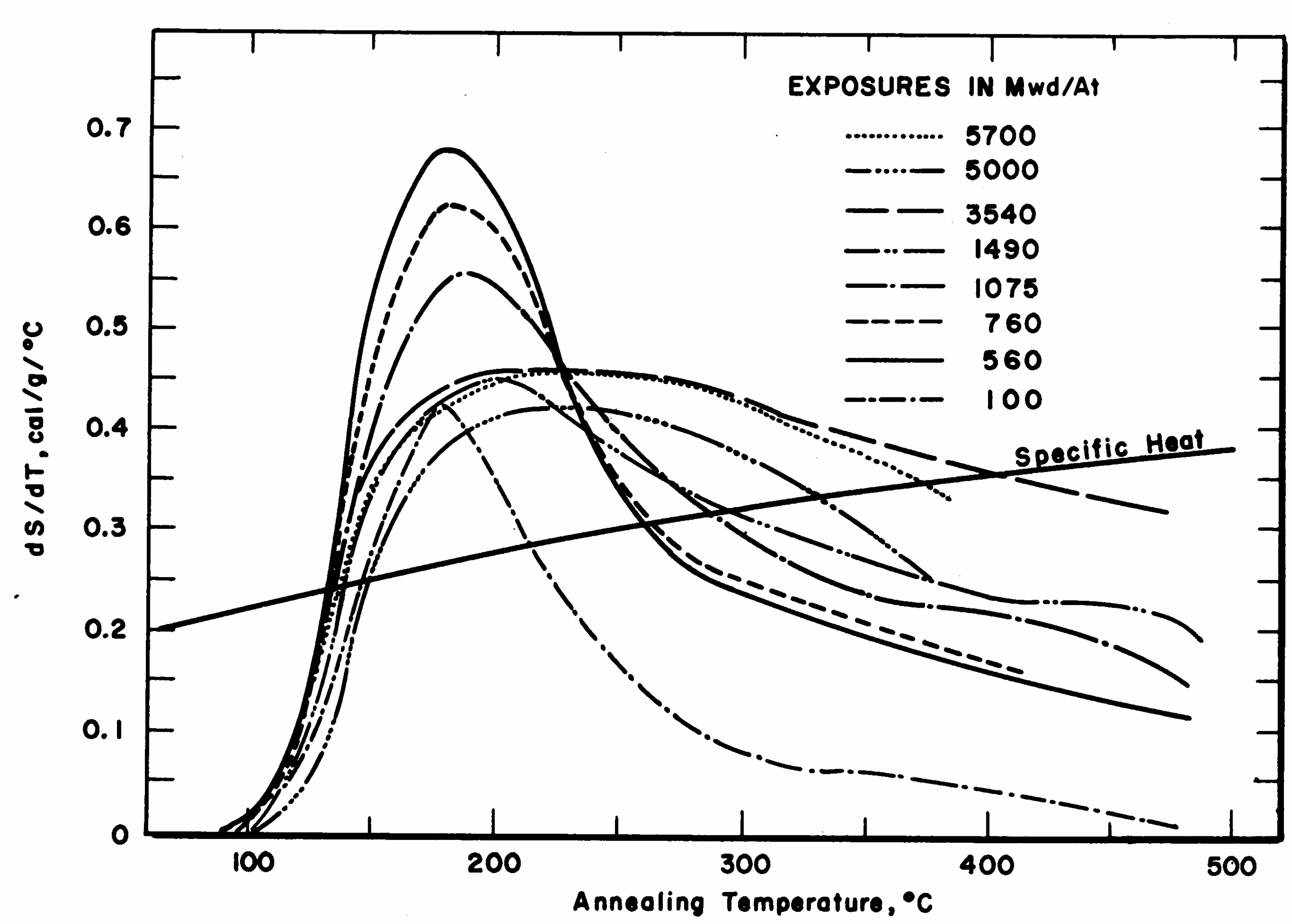


Figure 2.5 dS/dT curves for graphite irradiated near 30°C (from Nightingale 1965)

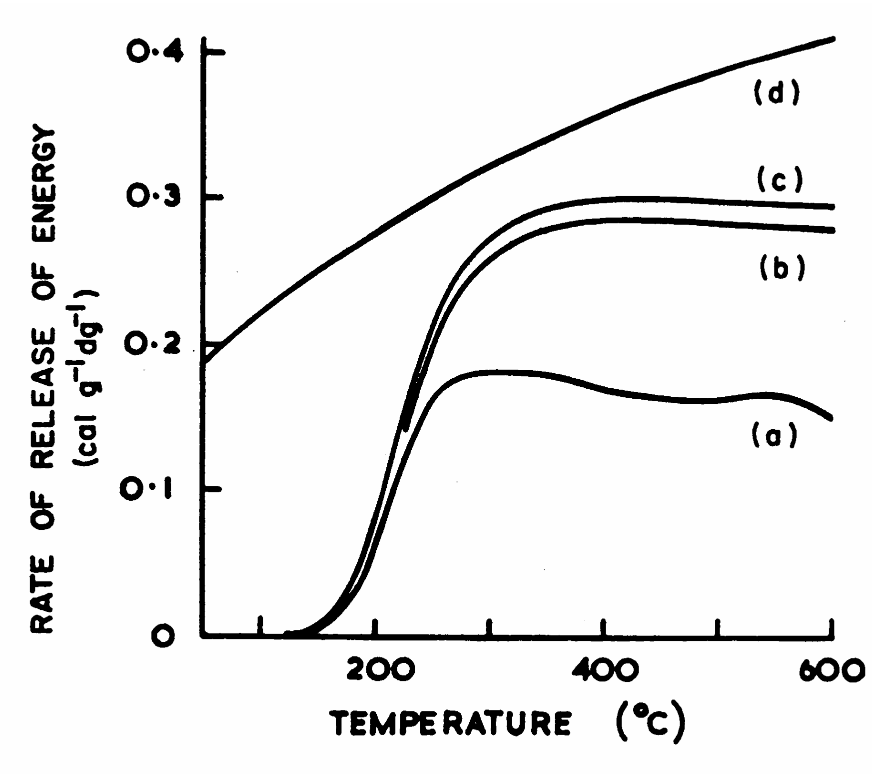


Figure 2.6 dS/dT curves for graphite irradiated at 150°C to different doses (a) 4.92×10^{20}, (b) 19.0×10^{20}, (c) 28.0×10^{20} n.cm^{-2} (d) Specific heat (from Simmons 1965)

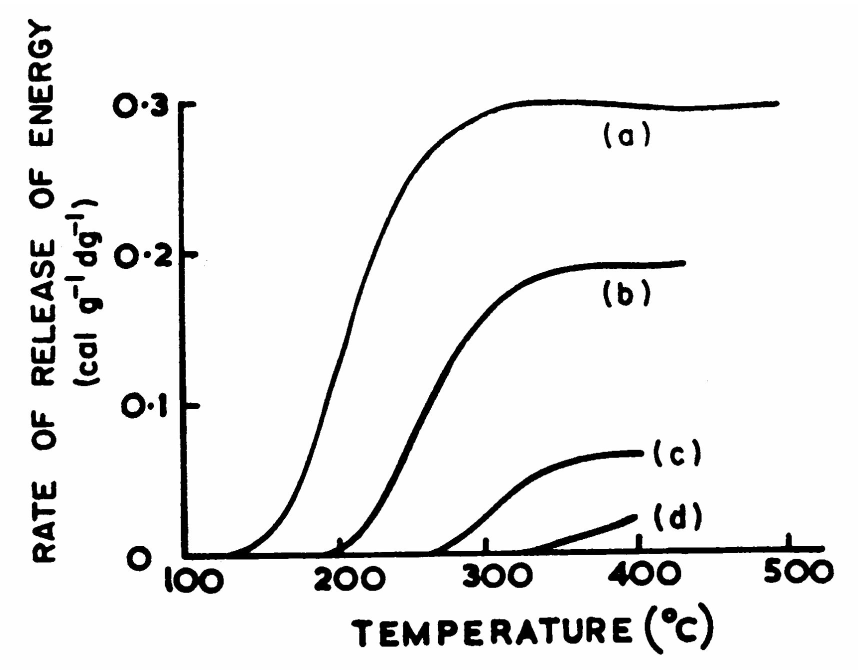


Figure 2.7 dS/dT curves for graphite irradiated at various temperatures (a) 150°C, (b) 200°C, (c) 250°C, (d) 300°C (from Simmons 1965)

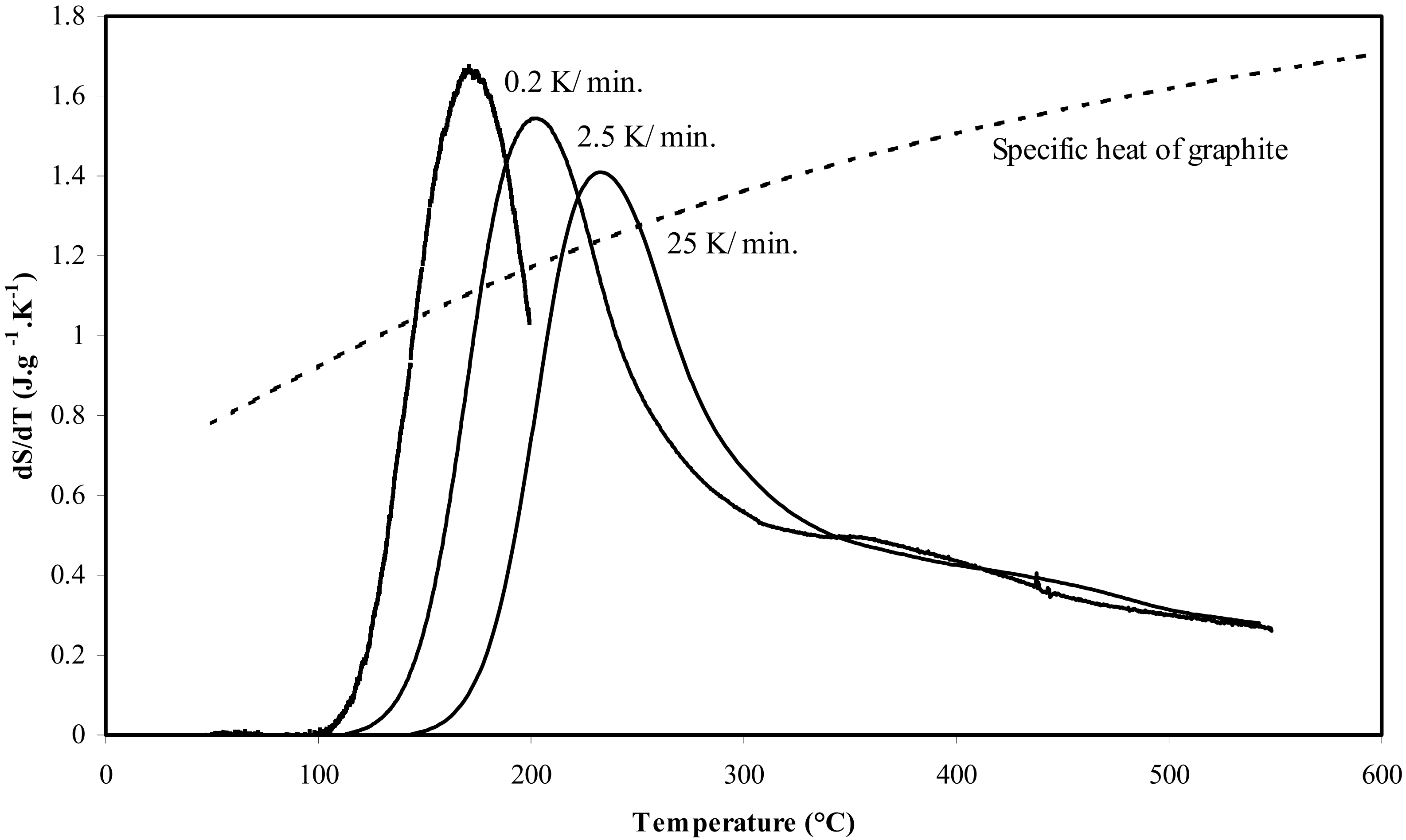


Figure 2.8 Adjacent samples measured at different heating rates

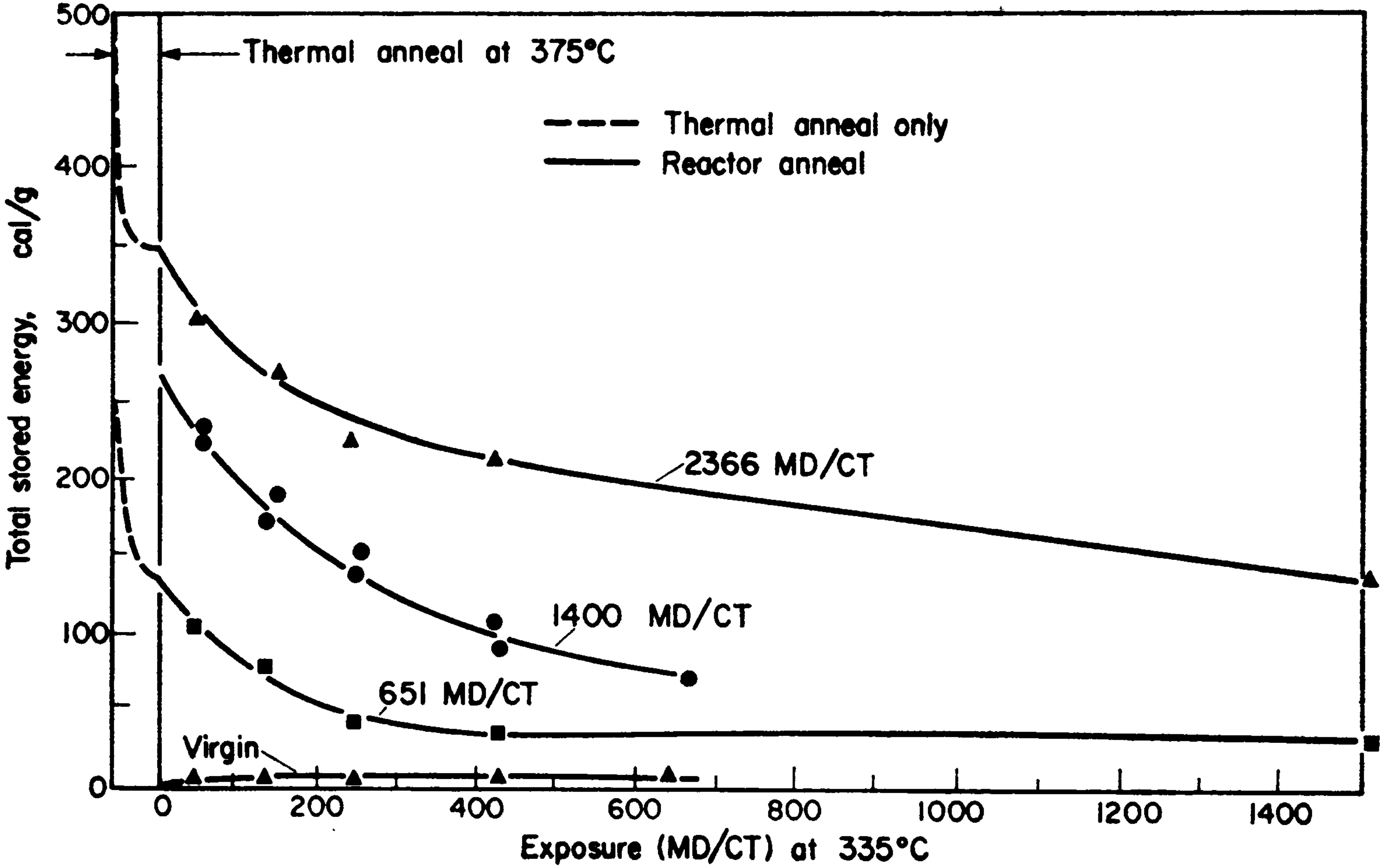


Figure 2.9 The effect of radiation annealing on the total stored energy in graphite irradiated at 30°C (from Simmons 1965)

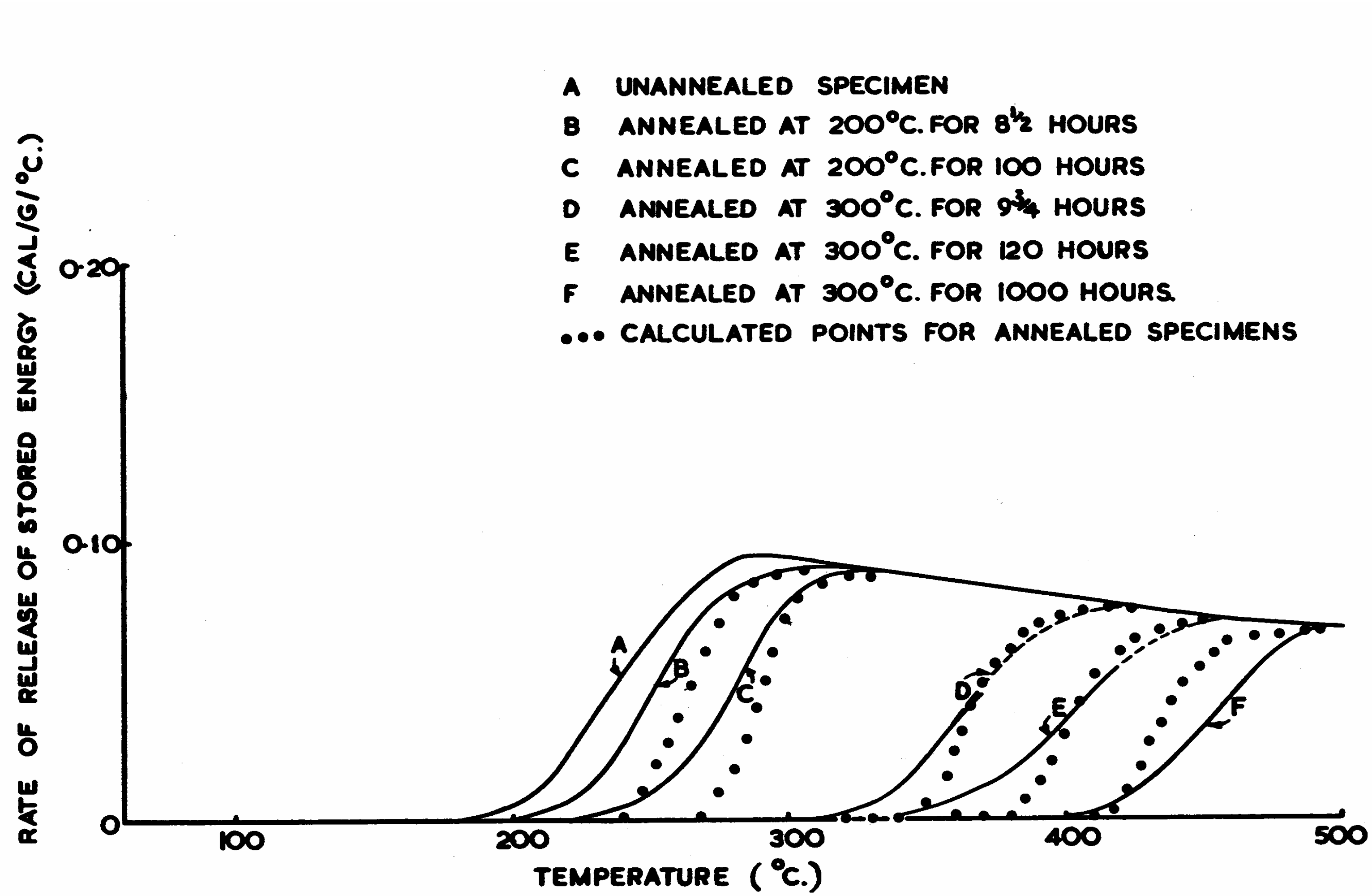


Figure 2.10 The effect of annealing at 200°C and 300°C for various times on the dS/dT curve of graphite irradiated at 155°C to a dose of 2.35×10^{20} n.cm^{-2} (from Simmons 1965)

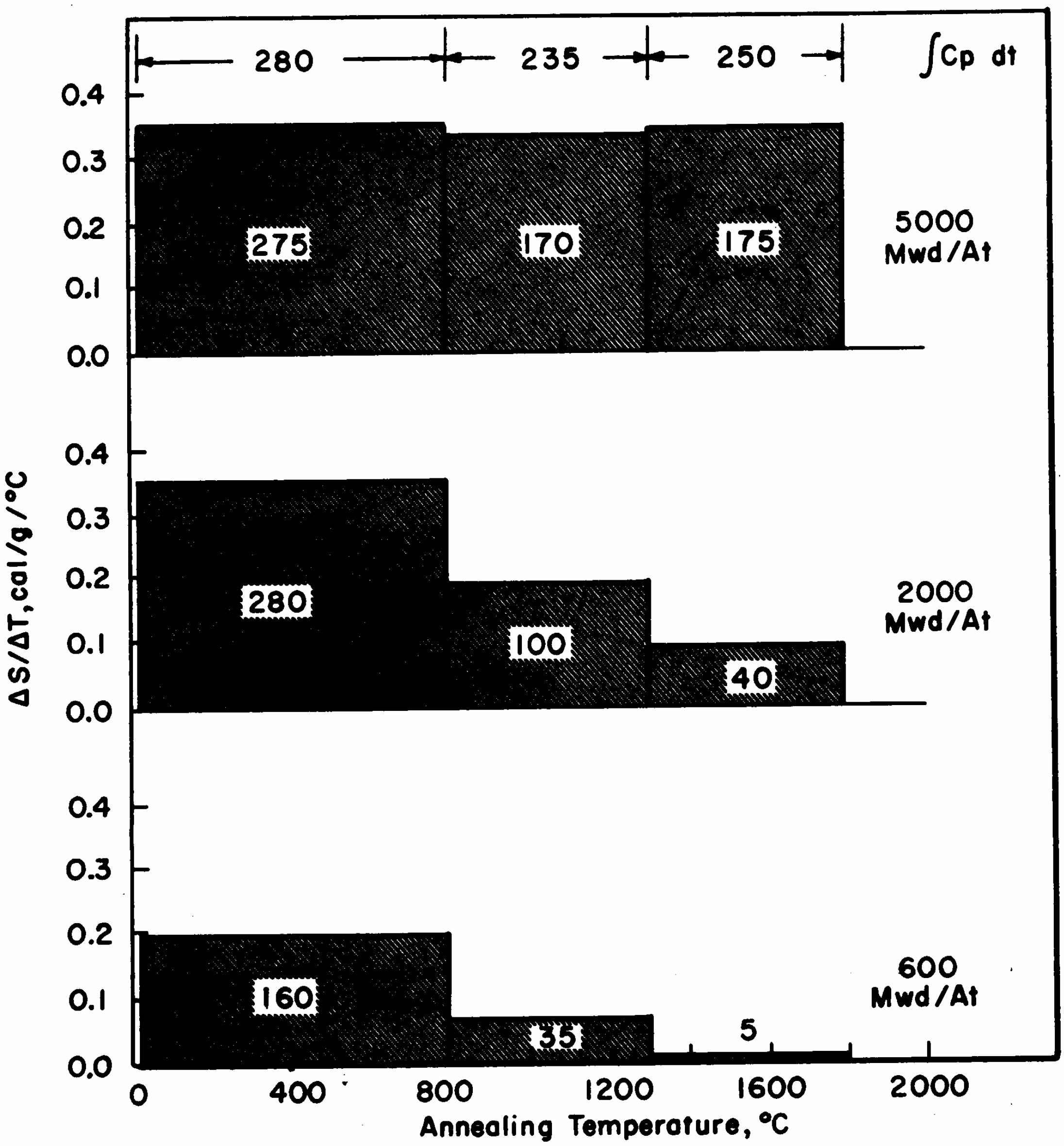


Figure 2.11 Stored energy released from graphite irradiated at 30°C. The numbers inside the areas give the stored energy released in the corresponding temperature range. The specific heat integrated over the temperature range is given at the top of the Figure. All stored energy was removed by a1800°C anneal (From Nightingale 1962)

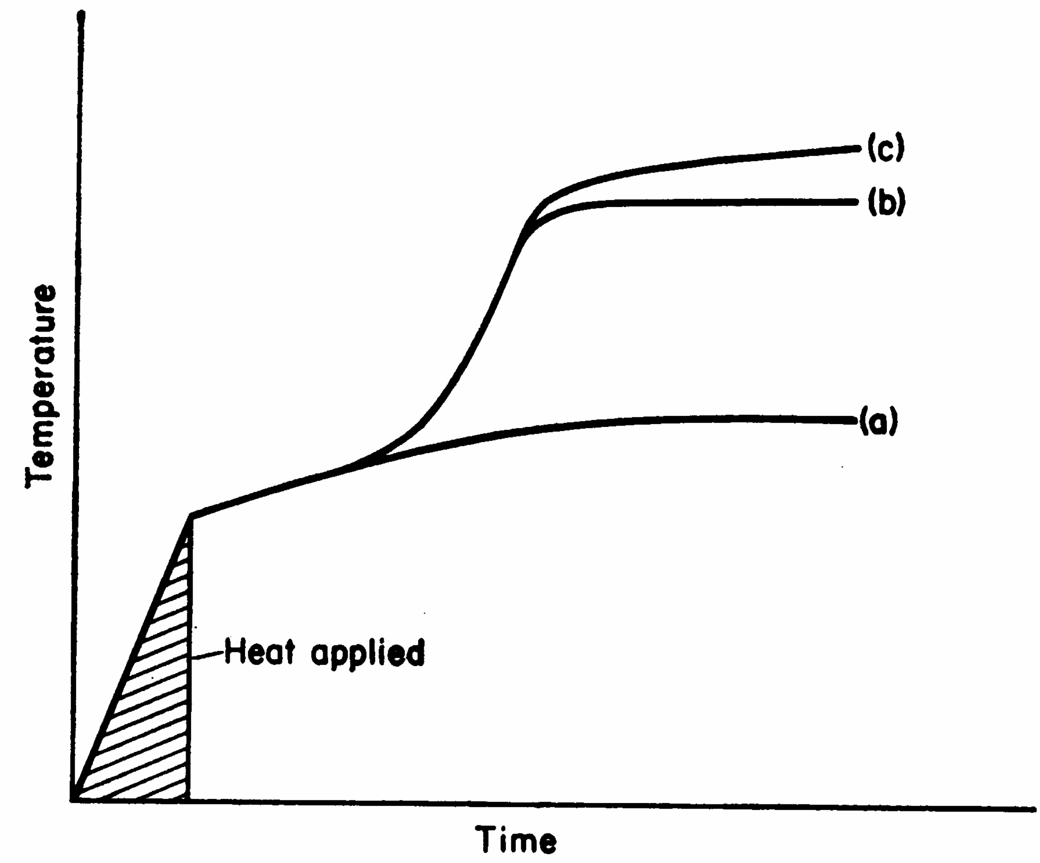
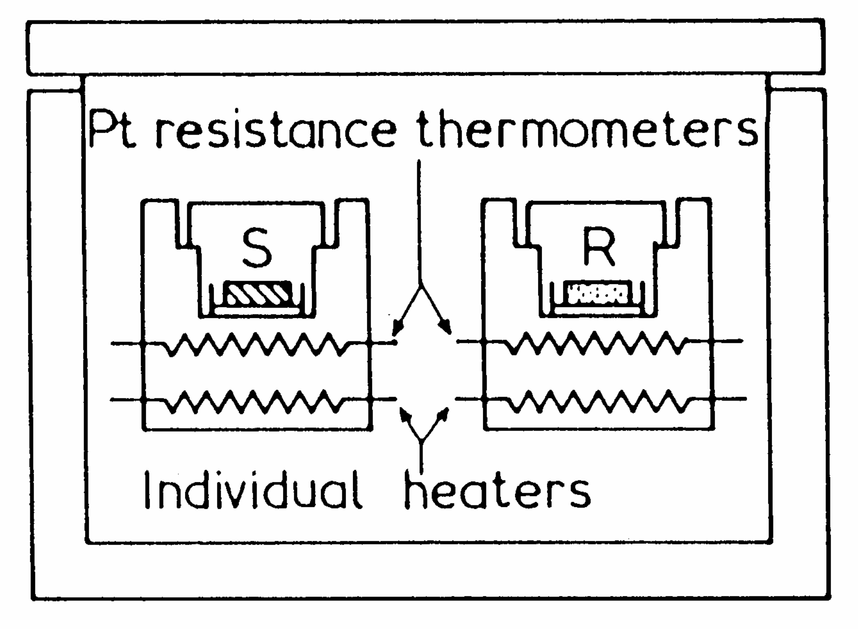
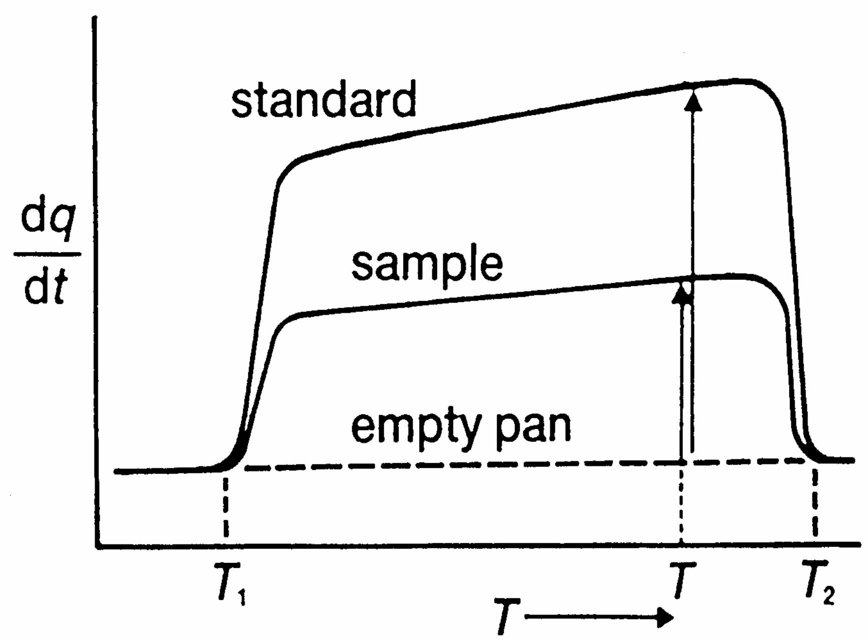


Figure 2.12 Typical adiabatic rise curves
(a) Low dose, (b) Moderate dose, (c) High dose (from Simmons 1965)



a) Schematic of the Perkin-Elmer DSC (from Compendium of Thermophysical Measuring Techniques, edited by Maglic K D, Cezairliyan A and Peletsky V E, Plenum Press 1984)



b) Specimen runs necessary to perform measurement (from Brown M E, Introduction to Thermal Analysis, Chapman and Hall 1988)

Figure 2.13 The linear rise calorimeter

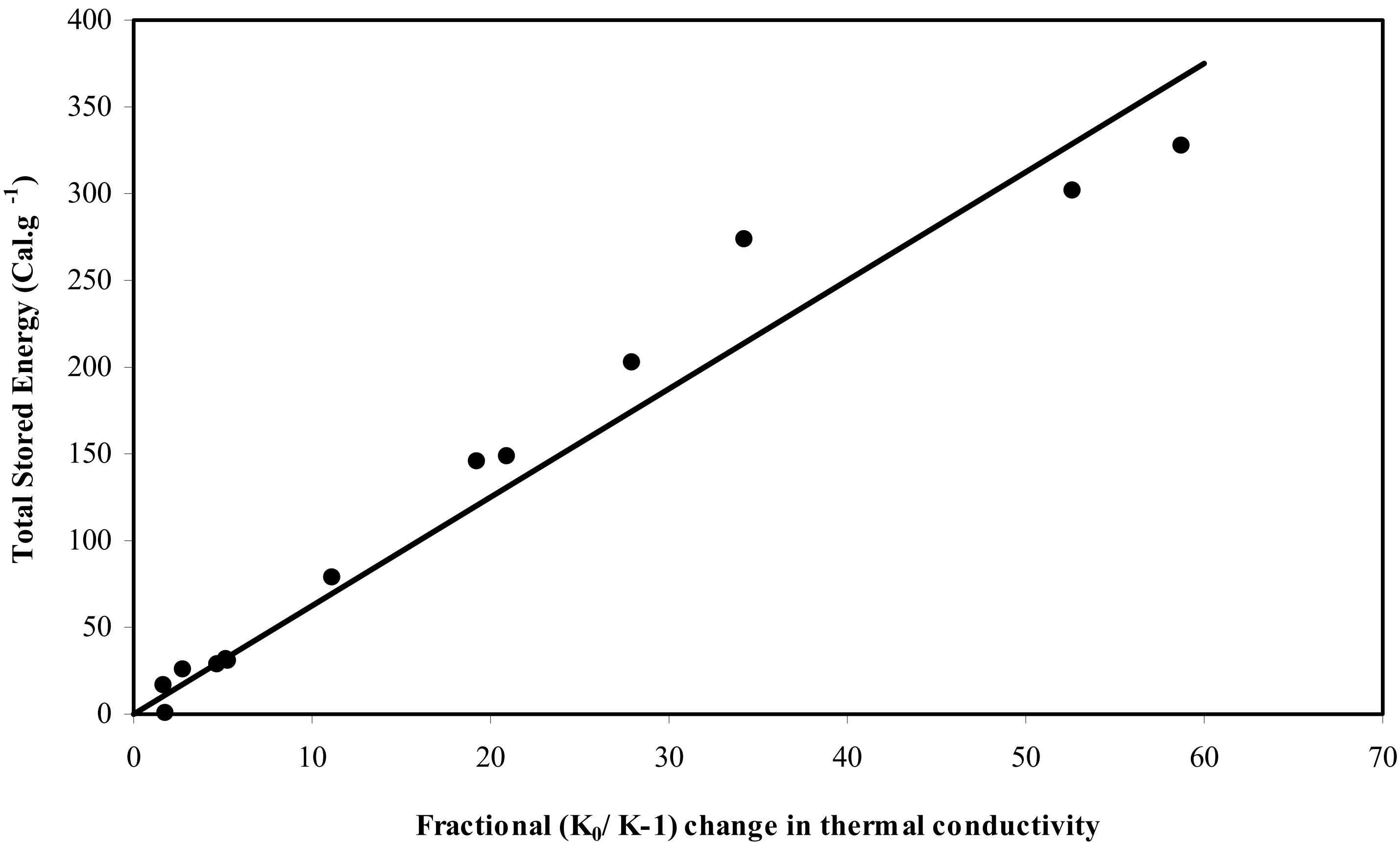


Figure 2.14 Total stored energy versus fractional change in thermal conductivity The maximum dose reached was 5.19 x 10^{21} n cm^{-2} (from Bell *et al*. 1962)

3. APPLICATION OF ASSAY TECHNIQUES

Direct measurement of the Wigner energy content of graphite in a reactor will probably be needed prior to decommissioning to determine if Wigner energy is an issue and later to provide quality control during waste processing. The need to obtain Wigner energy measurements before processing and the type and amount of process control will depend on the management route chosen, as shown in subsection 2.5 and Table 3.1.

Total stored energy measurements are unlikely to provide all the information needed for assessing management options. The information of interest is how much Wigner energy will be released if a piece of graphite is heated to a given temperature, at a given heating rate, and this is precisely the information obtained in a rate of release measurement using a differential scanning calorimeter (DSC, see subsection 2.2.3). Unfortunately there is no DSC currently available that can measure the rate of Wigner energy release at the very low heating rates expected in a deep waste repository. Consequently, the approach usually adopted is to measure the rates of release on small samples in a DSC at high heating rates, fit these to a variable activation energy model and then to use this model to extrapolate back to the low heating rates expected. This approach has been used in the validation and application of the Minshall model discussed in subsection 2.4. In the absence of any experimental data obtained at the correct heating rates, it is not possible to determine the uncertainty on these values. Experimental validation of the rate of Wigner release is desirable at the heating rates expected in a repository.

3.1 Process Control During Annealing

Rate of release measurements have already been made on graphite taken from the Windscale Piles to determine the effectiveness of annealing at various temperatures (Wörner, Botzem and Preston 2000). After each specimen had been annealed at a given temperature for three hours it was remeasured in a DSC and no further energy release was detected until the temperature of the graphite had exceeded the previous annealing temperature. The minimum annealing temperature should therefore be chosen to be above the maximum temperature that can be tolerated inside the waste package in a deep waste repository, including a margin of safety. The repository temperature concerned is in the region of 80ºC, as discussed in subsection 5.3.1.

The main difficulty with annealing is to ensure that all parts of the graphite have reached the minimum annealing temperature. This will need to be confirmed by adequate process monitoring during annealing (i.e. temperature measurement) with perhaps a confirmatory DSC measurement when the graphite has cooled. Given the measured surface temperature of the graphite, its thermal conductivity, its dimensions and time at temperature, it should be possible to calculate the minimum temperature reached anywhere in a graphite block.

It is unlikely to be practical to make multiple DSC measurements on all the graphite blocks after they have been annealed, and an effective sampling regime will need to be devised for quality checking purposes. Product sampling could be at its greatest during the commissioning of an annealing plant. A representative range of Wigner energy contents would need to be employed. From this the maximum level of residual Wigner energy would be confirmed, a value that could be fed into final assessments of package design. The uncertainties in the associated modelling would need to be taken into account. Systematic limited sampling might then be carried out for routine operation of the plant, although the ability to ensure that operating conditions permit all parts of the graphite blocks to reach the required temperature may provide sufficient assurance.

3.2 Process Control During Incineration/Steam Reforming

If the incineration or steam reforming disposal routes are to be followed, the Wigner energy content of the graphite is largely irrelevant in the sense that the graphite will be completely destroyed. However, some sampling of the waste will be needed to confirm that the process remains controllable during the release of Wigner energy in the plant.

3.3 Process Control During Direct Disposal

If graphite containing Wigner energy is to be disposed of by direct packaging, then thermal modelling must show that the package and contents must be able to withstand the ‘worst’ possible situation. Thermally inert material may need to be added to the package to reduce the maximum temperature rise (see subsection 2.7). Temperature monitors could be incorporated into the packaging to obtain early warning of Wigner energy release in disposal vaults before final closure.

Table 3.1 Potential requirements for Wigner energy measurements

Disposal route	Sampling regime for untreated graphite	Sampling regime for process control
Incineration	Possibly	No
Steam reforming	Possibly	No
Direct disposal	Yes	Not relevant
Partial annealing	Possibly	Yes

4. FACTORS AFFECTING WIGNER ENERGY RELEASE

The aim of this Section is to compile a comprehensive list of the events that could occur in a repository that might lead to the liberation of Wigner energy from irradiated graphite wastes and then to prioritise them by likelihood of occurrence. A word picture is developed of the sequence of events that could be involved in the liberation of Wigner energy. The range of impacts that such a release could lead to is discussed in Section 5.

It is assumed that the repository is located at a depth of some hundreds of metres and that packages are backfilled using a cementitious material. No other assumptions are made concerning the nature of the host geology or the design of the repository. The initiating event for Wigner energy release could occur after repository closure or a little before. Both timings could affect post-closure safety. The following subsections set out respectively the methodology employed; the initial set of events identified; and those of relatively high probability.

4.1 Methodology

The methodology employed is based on a comprehensive compilation by Nirex and AEA Technology of all the Features, Events and Processes (FEPs) that could occur in a deep repository. There are basically two types of FEP. So-called system FEPs are highly likely to occur (Locke and Bailey 1998), while probabilistic FEPs will only contribute to certain scenarios of how the repository may evolve (Billington and Bailey 1998). These compilations have been reviewed favourably at an international level by OECD/NEA (OECD/NEA 1999); they incorporate the NEA's own list of FEPs.

To assist in identifying the models that are required to simulate the development of a repository, small groups of FEPs are combined into conceptual models. The present analysis considers all the conceptual models and associated FEPs that have been derived to provide a complete description of the repository near field in the two references cited in the previous paragraph (see Table 4.1).

The entries in Table 4.1 have been used as a comprehensive checklist to determine which could lead to a rise in temperature in a repository. Those identified are listed in Table 4.2. The table notes whether each process identified has the potential to lead to a release of Wigner energy in the short term or in the long term. The probability of the event leading to Wigner energy release is set out in qualitative terms.

4.2 Initial Screening of Processes

Eight types of conceptual model or FEP have been identified that could lead to a temperature rise in a repository:

1. *Heat release associated with curing of the backfill.* Assuming that a cementitious backfill is put in place immediately before repository closure, it has been calculated that a peak temperature of about 60°C due to the curing exotherm would occur in waste packages a few days after the closure of the facility. This could lead to Wigner energy release on a short timescale (Guppy *et al.* 1999). This would occur towards the edge of a package: package centres were found to be insulated from this source of heat. The relative probability of this event leading to Wigner release is assessed as high.

2. *Heat release associated with radionuclide decay.* Radionuclides in a package containing irradiated graphite or in neighbouring packages could cause some rise in temperature as they decay. Modelling of this effect (Guppy *et al.* 1999) indicates a peak temperature of a little under 60 °C developing about 100 days into the post-closure period, with a slow decline thereafter to around ambient after perhaps 10,000 days (27 years). The temperature rise was found to be greater at the centre of the waste packages that at the edge, so this process could lead to the liberation of any remaining Wigner energy that could be released at relatively low temperatures.

3. *Degradation of metals in wastes and packaging and of organic materials by microbial attack.* These reactions could give rise to temperature increases in both the short and long terms. With potentially reactive metals in wastes (uranium, Magnox and aluminium), relatively rapid corrosion could occur when groundwater first contacts the metals following repository closure, with the new water composition disrupting the passivating layer. The timing of this will depend on the disposal geology selected and the quality of the packaging employed. Only part of the inventory of reactive metals will be affected at any one time, spreading the period of temperature rise and lowering the maximum value. Passivating layers are likely to reform relatively rapidly, slowing the rate of corrosion (Agg *et al.* 1996). Corrosion of the various steels in wastes and packaging will proceed slowly. Microbial degradation of organic wastes containing cellulose to give methane and carbon dioxide (Agg *et al.* 1996) could also lead to a temperature rise. These processes could be significant for the Sellafield mixed waste: it contains a wide range of metals and organic materials in close proximity to irradiated graphite. The degradable materials are less likely to lead to a widespread significant rise in temperature in graphite that is not packaged with them because of the thermal insulation provided by the intervening cementitious backfill. The timing of the temperature rise may lag behind that due to backfill curing and radionuclide decay, so there may by this time be little low temperature Wigner energy to be liberated. However, if levels of radioactivity are low in the packages, degradation processes may lead to release of Wigner energy in the short to long term.

4. *Seismic Events.* Seismic activity could in principle provide sufficient energy to initiate release of Wigner energy, but this should be an event of low probability because a repository location will be chosen in part for its low seismic activity.

5. *Magma.* Extrusion of magma into a repository from volcanic activity could provide sufficient heat to release Wigner energy in the long term. Again, the location for a repository will be chosen for its low activity in this respect. A low probability therefore attaches to this event.

6. *Human intrusion.* Intrusion into a repository could occur in the long term (beyond the period of institutional control) for exploitation of underground resources (water, oil, minerals, etc). The action of drilling for core samples could in principle initiate the release of Wigner energy. Given that a repository for intermediate-level wastes will be located at considerable depth (perhaps as deep as 1000 m), the probability of intrusion directly into a repository has been assessed as low (Billington and Bailey 1998).

7. *Criticality.* The probability of a criticality in a repository has been assessed as low (Nirex 2000). This includes scenarios involving the fissile material contents of individual packages as well as the possibility that fissile material from a number of packages could combine into a critical mass in the long term. If a criticality were to occur, the temperature rise would not necessarily be sufficient to initiate Wigner energy release, decreasing the probability of the latter still further.

8. *Meteorite impact that disrupts the repository.* It seems unlikely that this event would be other than of low probability, particularly given the considerable depth at which a repository would be located.

A further FEP was identified as being of relevance, heat transfer processes. This can be illustrated by the considerable impact that introducing a cement grout into a package of irradiated graphite has on decreasing the heat transferred from the surroundings, thereby slowing the liberation of Wigner energy (Guppy *et al.* 1999). In the cases modelled, the rise in temperature due to Wigner energy release was limited to the range 10-35°C, depending on the number of graphite-containing packages emplaced together. The maximum temperature reached was below 100°C. This relatively modest temperature rise is due to the dilution of the graphite in the packages and the heat sink provided by the grout. When the graphite occupied the entire package (i.e. no grout was present), the temperature rose quickly to 140°C, and was still increasing rapidly at this point. In the same way, the backfill surrounding each package should also decrease appreciably the heat that can be transferred into a package of graphite from neighbouring ones. The metals and organic materials present in the mixed waste at Sellafield may also provide a heat sink that can slow the rate of release of Wigner energy.

A further effect is the possibility that the γ-irradiation field in a repository may increase the amount of Wigner energy that can be released at low temperatures. The reader is referred to subsection 2.7, where the current uncertainties associated with this possibility are discussed. It is not considered further here because the probability associated with the effect is assessed as being low, although the impact of such an event could be considerable. Further study of the issue is recommended to assess the significance of this area of risk.

4.3 Identification of Priority Processes

The analysis in the previous subsection indicates that there are three processes of relatively high probability that could lead to the release of Wigner energy. The sequence of events could be as follows:

- Heat release associated with curing of the backfill would affect all the repository in the very short term, and could be transmitted directly into packages filled with graphite, with a subsequent temperature rise due to release of Wigner energy (Guppy *et al.* 1999).
- Heat released as a result of radionuclide decay could release Wigner energy on a slightly longer timescale: energy stored towards the centre of packages that was not liberated during curing of the cement might be released.
- Various reactions involving metals and organic wastes (corrosion and microbial degradation) could be significant in the longer term in releasing Wigner energy if initial levels of radioactivity are low, particularly towards the centre of packages. This could be significant in packages containing the mixed waste in storage at Sellafield.

These processes are the same as those identified in earlier work (Guppy *et al.* 1999), but with the advantage that they arise from consideration of a comprehensive list of FEPs that could occur in a repository.

These processes could lead to substantial increases in temperature if the effect of the initial heat source is not diluted by a thermal sink provided by a cementitious encapsulation grout or the backfill, or, in the case of the mixed waste at Sellafield, other solids in the waste.

It is recommended that current modelling of temperature rises in packages containing graphite wastes (e.g. Guppy *et al.* 1999) be extended to (a) the mixed waste and (b) the effect on immediately neighbouring packages.

Table 4.1. Checklist of near-field FEPs and conceptual models used to identify processes that could lead to release of Wigner energy

Notes: System CMs and FEPs from Locke and Bailey, 1998.
Probabilistic FEPs from Billington and Bailey, 1998.
(1-8) after an entry denotes a CM or FEP that could potentially lead to release of Wigner energy. The numbers refer to the processes discussed in subsection 4.2.
(×) denotes a CM or FEP that would not be involved in the release of Wigner energy.

Conceptual Model (CM)	**Relevant Features, Events and Processes (FEPs) and Keywords**
System CMs and FEPs	
Inventory of radionuclides in waste at closure (2) Chemical composition of engineered system at closure (3) Physical conditions at closure (1,2,3) Composition and distribution of gas trapped in engineered system at closure (×)	Radionuclide inventory (2) Ventilation (during operations) (×) Repository design (1,2,3,7) Packaging and containers (3) Waste materials composition (3) Pathways (cracks, pores) (×) Backfill strategy (1) Waste emplacement strategy (2,7)
Thermal evolution (1,2,3) Engineered system characteristics: environmental pressure (×) Microbial processes in engineered system (3) Redox evolution (×) Degradation of organic materials (3) pH evolution (3) Container failure (×) Corrosion (3) Resaturation (3) Effects of radiolysis (2) Quantity of bulk gas in engineered system (×) Transport pathways in engineered system (×) Determination of gas pathways (×)	Heat generation (cement hydration, radiogenic) (1,2) Heat transfer processes (All) Chemical degradation (3) Gas generation (microbial, chemical, corrosion) (3) Container corrosion (aerobic, anaerobic, galvanic, stress -related, localised) (3) Radiolysis (2) Internal stress (×) Resaturation (3)

Conceptual Model	Relevant FEPs and Keywords
Flux of radionuclides from engineered system: gas phase (×) Flux of radionuclides from engineered system: liquid phase (×) Speciation of ions and radionuclides (×) Sorption (×) Formation and presence of colloids (×) Solubility of radionuclides in liquids (×) Transport of radionuclides in aqueous phase liquid in engineered system (×) Radionuclide decay and ingrowth (2) Concentration of radionuclides in bulk gas in engineered system (×) Gas-driven water flow (×) Dissolution/precipitation (×) Mineralisation: crystallisation (×) Transport of gas through bulk solids (×)	Chemical & physical form of wastes (3) Groundwater chemistry (e.g. sorption, complexation (×) Driving forces (e.g. pressure gradients, gravitational forces) (×) Dissolution of radionuclides (×) Liquid phase transport (advection, dispersion, molecular diffusion, & rock-matrix diffusion) (×) Gas generation (e.g. by corrosion, microbial degradation of organics) (3) Resaturation (3) Gas phase transport (continuous or bubble phase) (×) Radioactive decay and ingrowth (2) Exchange between liquids, solids and gas (×) Dissolution (×) Precipitation (×) Solubility (×) Sorption (×) Mineralisation (×) Isotopic exchange/dilution (×) Exclusion (×)
Probabilistic FEPs	
Wells and boreholes for resource exploitation (6) [1]	A large number of FEPs concerning abstraction of water, gas, minerals and brine and associated exploratory activities (6)
Criticality incidents (7)	Criticality in engineered system (7)
Disruption of geosphere and engineered system (4, 5, 8)	Magma: deep intrusion (×), extrusion (5) Meteorite impact: repository disruptive (8)/non-disruptive (×) Seismic events (4)

[1] The entries in the left-hand column for probabilistic FEPs are generalised FEPs rather than Conceptual Models. The right-hand column gives a more detailed account of each.

Table 4.2. Screening of FEPs that could potentially lead to release of Wigner energy in a repository

	(S)hort or (L)ong-term Release	Relevance to Wigner Energy Release	Probability of Wigner Energy Release
1. Backfill strategy; Heat generation (cement hydration, radiogenic) (S)	S	Curing exotherm for backfill may supply sufficient energy to initiate Wigner release	High
2. Radionuclide inventory; Radiolysis; Radionuclide decay and in-growth; Effects of radiolysis (S)	S	Radionuclide decay heat could be supplied to irradiated graphite. Could be low by the time a repository is closed, particularly if there is an extended period of care and maintenance	High
(-)Heat transfer processes (S)	S/L	Cement grouts and backfills good thermal insulators	Important in controlling temperature rise due to Wigner energy release
3. Gas generation (microbial, chemical, corrosion); Corrosion; Degradation of organic materials; Container corrosion; Chemical degradation; Chemical composition of system; Packaging & containers; Resaturation; Microbial processes (S)	S/L - corrosion of metals, microbial degradation of organics	Reactive metals will have passivating layers at repository closure. Rapid gas evolution could occur for a time as groundwater enters repository, but event will be spread in time. Localised environments could exist that encourage microbial and corrosion 'hotspots'.	High - particularly when processes occur in packages containing irradiated graphite and/or radioactivity levels are low.
4. Seismic events (P)	L	Repository site will be selected in part for low seismic activity	Low
5. Magma - extrusion (P)	L	Site selected in part for low volcanic activity	Low
6. Human intrusion: wells and boreholes for resource exploitation (P)	L - beyond period of institutional control of repository site	Probability low that intrusion would be as deep as the repository itself	Low
7. Criticality in engineered system (P)	L	Repository design and fissile limits on packages limit probability of a criticality to low levels	Low
8. Meteorite impact: repository disrupted (P)	L	Impact with repository disruption low probability	Low

Note: (S) denotes system CM or FEP, (P) a probabilistic FEP.

5. POTENTIAL IMPACTS OF WIGNER ENERGY RELEASE IN A REPOSITORY

The objective of this Section is to construct a methodology for examining the effects of Wigner energy release on the long-term safety of a repository. Some illustrative assessments are performed.

5.1 Post-closure Safety in Deep Repository

Long-term safety in a sealed, deep repository is determined by the physical and chemical properties of the barriers in the system that separate the emplaced waste from man. Working outwards from the waste, they are:

1. Any encapsulating material used to immobilise the waste. Various cements are commonly used in the UK for this purpose. As an alternative, some wastes are compacted into containers, often with an annulus of grout rather than being intimately grouted. In both cases, the product is known as the wasteform.
2. The packaging that contains the wasteform. This can be made of mild or stainless steel or concrete.
3. The backfill used to surround each package and create a monolithic mass in the disposal vaults. In the UK, a cementitious backfill is proposed (Francis, Cather and Crossland 1997). The high pH exerted by this type of backfill helps to restrict the solubility of many radionuclides and also provides sites for the sorption of radioactive species in a dissolved state.
4. The host rock that contains the repository. A very low rate of water flow through the host rock is an important criterion on which the repository site will be selected. Additional protection is afforded by using a considerable depth for disposal of up to 1000 m.

Currently the nature of the disposal geology to be used in the UK is uncertain, and the latest assessment of post-closure safety is generic to a range of geologies (Nirex 2000). For this reason, the methodology presented here focuses on the safety performance of the repository near field (items 1-3 above) and does not take into account the additional protection afforded by the host rock. This approach has three benefits:

- The extent to which the design intent of the repository has been fulfilled with respect to post-closure safety can be assessed.
- An approach based on near-field safety may be easier for a wide audience to appreciate than the necessarily more complex full assessments that trace the movement of radionuclides through the host rock and biosphere to man.
- It is robust in terms of present uncertainties over the setting for a repository.

The physical integrity of the near-field barriers, and hence the degree to which radionuclide movement is prevented, is designed to extend over a few hundred years (Chambers *et al.* 1995). Beyond this, the containers may fail progressively due to corrosion (metal) or cracking (concrete) and the backfill may also become cracked. In addition, vents and corrosion pinholes may admit groundwater. The period of barrier integrity is designed to accommodate the radioactive decay of isotopes with half-lives up to about 30 years. Isotopes of this type that are often found in wastes include ^{60}Co (half-life about 5 years) and ^{90}Sr and ^{137}Cs (both 30 years). The latter pair in particular are expected to dissolve relatively readily in repository waters, and so physical barriers that accommodate about 10 half-lives of decay are an important aspect of post-closure safety.

For longer-lived radionuclides, the approach to safety is based on achieving chemical control over aqueous concentrations of radionuclides (Chambers *et al.* 1995). This is obtained through:

- Control of long-term pH of groundwater to high values by the cementitious backfill (10-12).
- A reducing environment caused by metals and microbial processes that rapidly consume the oxygen trapped in the repository at closure. The resulting low oxidation states are usually less soluble in water than high oxidation states.
- Sorption sites for many radioelements provided by the cementitious backfill and the corrosion products of the metals (particularly steels).

These features of repository chemistry limit aqueous concentrations of a wide range of fission and activation products and actinides to low levels.

A further feature of near-field performance is gas generation (Agg *et al.* 1996). Corrosion of the metals present in wastes and packaging will generate hydrogen. Waste Magnox, aluminium and uranium are reactive metals that could give rise to hydrogen relatively rapidly, though the formation of passivating layers usually limits the duration of this behaviour. Steels and Zircaloy in wastes and packaging corrode more slowly. For steels and uranium, the oxygen trapped in the disposal vaults at closure must be consumed before hydrogen generation occurs. Magnox, aluminium and Zircaloy corrode to give hydrogen irrespective of the presence of oxygen.

The corrosion rates of steels, Zircaloy and Magnox are minimised at high pH, but the rate for aluminium increases as pH is increased from 7 to 12 (Baker *et al.* 1997). The rate of corrosion of uranium is essentially insensitive to pH in the range 2 to 14 (Baker *et al.* 1966).

In addition, attack by microbes on organic wastes containing cellulose leads to the formation of carbon dioxide and methane under the reducing conditions expected in a repository. The high pH in a repository is expected to minimise the rate of gas formation by this route (Rees 1989).

A repository needs to be designed so as to permit the gas generated to move readily through the near field and disperse in the host geology without leading to pressurisation effects that could damage the integrity of near-field barriers and the surrounding rock. Current assessments indicate that this is achievable (Nirex 2000).

A number of indicators of near-field performance are defined in subsection 5.2. The effect of Wigner energy release on these indicators is examined in subsection 5.3 with respect to two of the temperature profiles obtained in recent modelling of packages containing graphite (Guppy *et al.* 1999). The level of risk associated with the perturbations to repository safety caused by Wigner energy release is examined in qualitative terms in subsection 5.4.

5.2 Indicators of Near-field Safety

Five indicators have been selected for the methodology for assessing packages containing irradiated graphite:

1. Physical integrity of near-field barriers so as to contain radionuclides of half-life up to 30 years.
2. Chemical control over concentrations of longer-lived radionuclides in repository waters.
3. Short-lived effects on the groundwater pathway due to the transient changes in temperature brought about by Wigner energy release.
4. Long-term effects on the groundwater pathway due to Wigner energy release.
5. The amount of gas leaving the near field.

These are now discussed in terms of two temperature profiles due to Wigner energy release obtained in recent modelling (Guppy *et al.* 1999). Two extremes in the results reported in that paper are considered. They have been selected to illustrate the recommended methodology for assessing risk due to Wigner energy release; the results obtained might be revised in the light of details of the package proposed and consideration of the degree of confidence attaching to the modelling (see Section 3). The ambient temperature in the repository was assumed to be 30°C. The scenarios are as follows:

1. A 4 m box containing 100% 'worst case' irradiated graphite (so maximising Wigner energy release) that is surrounded by cemented wasteforms containing no irradiated graphite. The curing exotherm for the backfill triggers a rapid increase in temperature due to Wigner energy release that is still increasing strongly when the modelling run terminates at about 140°C.

2. As in 1. above, but with the 4 m box containing a 50:50 mixture of 'worst case' graphite and cement grout arranged evenly throughout the container. Here, the maximum temperature from Wigner energy release is limited to 78°C at the edge of the package. The maximum at the centre is modelled to be 65°C. The grout acts as a heat sink for much of the heat of curing of the backfill, leading to a more controlled release of Wigner energy. The difference in temperature between the surface and centre of the box arises from the higher temperature due to backfill curing at the edge of the box compared with its centre (see below). The less even the distribution of graphite in the box, the higher the temperature the waste reaches.

These temperatures should be seen in the context of background temperatures in the packages due to curing of the backfill ranging from about 50°C at the centre to 60°C at the edge.

5.3 Assessment of Effects of Wigner Energy Release

5.3.1 Scenario with maximum temperature 78°C

Performance of near-field barriers. The cementitious backfill preferred by Nirex - the Nirex Reference Vault Backfill (NRVB) (Francis *et al.* 1997) - is designed to accommodate vault temperatures up to about 80°C (Nirex 1995). The effect of such transient temperatures on the physical condition of the backfill and its chemical properties (pH, redox potential and sorption capacity) are believed to be minor in terms of repository safety (Nirex 1995 and Goldberg *et al.* 1997).

The formation of acidic hydrogen chloride from waste polyvinyl chloride (PVC) may be accelerated by an increase in temperature (Goldberg *et al.* 1997), but the amount of backfill present should absorb it readily (with calcium chloride being formed). Cellulose in wastes can degrade to give water-soluble complexing agents for actinides, notably for plutonium. This can increase their solubility in repository waters. An increase in temperature may increase the rate at which these agents form (Goldberg *et al.* 1997), but the effect should be limited because:

- The temperature rise in the graphite will be appreciably less when it is transmitted to a surrounding package containing organic wastes because of the heat sinks provided by the surrounding backfill and any grout in the latter.
- The timescale of the temperature transient is modelled to be brief in duration, at around 100 days (Guppy *et al.* 1999).

Thus the effect of this temperature excursion on the four performance indicators concerned with the concentration of radionuclides in repository water and their flux on leaving the near field is essentially insignificant.

Gas generation. A maximum increase in temperature of 20°C at the edge of a package containing irradiated graphite could increase the overall rate of gas production due to corrosion by causing a knock-on rise in temperature in a neighbouring package.

The repository is likely to be still aerobic given the short timescale of the Wigner energy release in this scenario. The corrosion of steels may be accelerated during the short period the temperature transient lasts (about 100 days), but the process produces no gas. The corrosion of uranium could be accelerated substantially during this period, but again the aerobic process produces no gas. The corrosion rate of uranium in air saturated water increases by a factor of 80 between 60°C and 80°C (Waber 1952). A mitigating factor concerns the availability of the water needed to support corrosion shortly after repository closure. Groundwater will not be able to access the wastes at this stage and any free water initially in grouted packages may have been consumed before repository closure.

Corrosion rates for Magnox increase with temperature (Bradford *et al.* 1978). Above 80°C, the corrosion layer breaks away, increasing the rate of corrosion. In the present instance, the maximum temperature is below 80°C. The corrosion rate for pre-breakaway corrosion is given by:

Rate = $1.2\ 10^{15} \exp(-10720/T)$ µm/year.

Increasing the temperature from 60°C to 78°C at the edge of a package increases the rate of corrosion of Magnox from 12 to 65 µm/year at around pH 12. However, the limited duration of the transient, the dissipation of heat in the backfill, and the lack of access for water to the metal shortly after repository closure suggest that the impact on the gas pathway will be minor.

The rate of corrosion of aluminium in alkaline media also increases with temperature. In 80-day experiments, the rate was found to increase by a factor of about 5 between 30°C and 60°C at pH 11 (Tabrizi *et al.* 1991). However, the limitations noted on Magnox corrosion soon after repository closure would be expected to apply to aluminium.

Metals that derive from power reactors (including parts of the inventories of steel, Zircaloy and Magnox) contain tritium that may be liberated as tritiated hydrogen as corrosion proceeds (Baker *et al.* 1997). This gas can also be liberated by the diffusive movement of tritium out of the metal, the rate of which will increase with temperature. The corrosion of uranium can lead to the formation of small amounts of methane, presumably due to a carbide impurity in the metal (Baker *et al.* 1966). Corrosion of irradiated uranium may therefore lead to the formation of $^{14}CH_4$. Increases in corrosion rates could therefore increase the rate of formation of radioactive gases.

In contrast, the metabolism of microbes may be prevented or substantially suppressed for much of the time the temperature remains above about 50°C. Microbes can survive brief exposure to high temperatures, and their activity may be restored when conditions are more favourable (Nirex 1995). Microbial gas production may thus cease during the temperature transient, but restart later.

Summary. The performance of the repository is unlikely to be significantly degraded with respect to the groundwater pathway by a steady release of Wigner energy. This conclusion may extend to the gas pathway, but it would be prudent to confirm this in assessments of the knock-on temperature increases in neighbouring packages due to Wigner release, the implications this has for gas generation from metals (including the production of tritiated hydrogen and gas containing ^{14}C), and whether transients in gas production could physically disrupt the engineered near field.

Mitigating features of the system with respect to gas production include:

- A possible low availability of free water immediately after repository closure.
- A temperature rise in a neighbouring package that would be less than the 20°C rise obtaining at the edge of the graphite package.
- A temperature transient that would cease to exist about 100 days following closure.

Although the repository system may be able to accommodate temperature transients of more than 80°C, a restriction appears at 100°C due to water boiling. A design target of 80°C therefore includes a safety margin, and it is not recommended that this margin be decreased to accommodate Wigner energy release.

5.3.2 Scenario with a rapid rise in temperature extending above 140°C

This scenario is also initiated shortly after closure by the curing exotherm of the backfill. The lack of a heat sink in the 4 m box gives a much less controlled release of Wigner energy. The duration of the transient is unknown.

Physical integrity of near-field barriers. On this scenario, widespread degradation of near-field barriers might be expected in the package of graphite itself and in surrounding backfill and packages:

- The acceleration in corrosion rates will affect reactive metals in neighbouring packages and mild steel containers and wastes. Rates would increase up to 100°C, beyond which vapour phase corrosion would occur. The latter may be at a lower rate than the former due to limitations on the availability of water.
- The volume increases accompanying corrosion might lead to cracking in the in-package grouts and could also affect the integrity of containers and the backfill.
- Thermal expansion of metals in wastes and packaging may also lead to some loss in physical integrity.
- The temperature excursion involved in this scenario (and therefore the thermal gradient set up) is likely to be high enough to lead to cracking in the backfill in its own right. Concrete containers may be similarly affected.
- Increases in the rate of gas formation could generate sufficiently high local overpressures as to damage near-field barriers. Conversion of water to steam could contribute to this, depending on the availability of water.

Overall, the degree of protection afforded to short-lived and relatively soluble isotopes by the near-field barriers could decline significantly. Extensive cracking could increase the rate of water movement through the repository, leading to a permanent increase in the flux of all radionuclides from the engineered system.

Chemical protection due to near-field barriers. A number of effects could occur at high temperatures that would, if realised, remove at least some of the chemical features that restrict the solubilities of a range of long-lived radionuclides to low values:

- A transient increase in temperature could lead to transient increases in the solubilities of a range of radionucludes.
- Cracking of the backfill would concentrate groundwater flow along the cracks. The species that maintain pH at a high value (including calcium hydroxide) would be depleted in the region around the crack, and pH could fall (Harris *et al*. 1997).
- Increasing the temperature up to 200°C gradually leads to the formation of a range of minerals in cements and the consumption of amorphous calcium silicate gel (Atkinson *et al*. 1995). Above 100°C, these minerals include tobermorite, xonotlite, portlandite and gyrolite. Some of these minerals buffer pH to values below 10, which could cause radionuclide solubilities to rise. However, cements with a high calcium:silicon ratio, such as NRVB, mineralise to include portlandite, or calcium hydroxide. This mineral buffers pH to about 12. Temperatures significantly above 200°C could lead to consumption of calcium hydroxide, and a loss of control over pH to high values. In-package grouts will probably have lower calcium:silicon ratios than NRVB, which could lead to some loss of pH buffering capacity approaching 200°C.
- Formation of minerals could decrease the surface area of the various cements present. This could be beneficial in reducing the extent of crack formation or deleterious in decreasing the extent of sorption of radionuclides (Goldberg *et al.* 1997).
- The comments above concerning mineral formation are based on the behaviour of 100% cements. No account has been taken of reactions between waste materials and the various cementitious materials present. These processes could affect radionuclide solubilities and sorption.

In contrast, the increased rates of corrosion of steel and uranium accompanying the temperature increase could lead to reducing conditions being established more rapidly. This could lead to a decrease in the aqueous concentrations of many radionuclides in the short term.

Overall, a number of effects could occur during a temperature transient well above 140°C that could modify radionuclide solubility and sorption and groundwater flow paths. Eventually around 5,000 te of low-temperature irradiated graphite may be disposed to a deep repository (see subsection 2.5). The effects of Wigner energy release may spread to neighbouring packages. The flux of radionuclides from the repository as a whole could be affected significantly should a large part of the repository become involved.

Gas generation. As noted earlier, increasing temperature increases gas generation by corrosion up to 100°C. This could include an acceleration in the production of tritiated hydrogen. Oxygen trapped in the repository at closure could be consumed more rapidly, leading to a low redox potential being developed sooner. Hydrogen production from steel and uranium would then start earlier. The hydrogen formed would be joined in the gas phase by water vapour above the boiling point of water. Localised pressurisation and possible damage to near-field components could ensue. Microbial action to produce gas will cease at high temperatures, and it is possible that parts of the repository will be sterilised. Microbes are likely to re-enter the repository in groundwater when the temperature subsequently decreases towards ambient

Summary. The analysis above suggests that all five indicators of repository safety could be affected adversely in the scenario where the maximum temperature reached is well above 100°C:

- Containment of short-lived radionuclides by the engineered barriers may be degraded.
- Chemical control (solubility, sorption) over concentrations of long-lived radionuclides in groundwater may be impaired.
- Transient effects in the groundwater pathway may occur, e.g. temperature rise increasing radionuclide solubilities for its duration.
- Significant long-term effects in the groundwater pathway may arise due to some loss of control over radionuclide chemistry and the development of new flowpaths for groundwater through induced cracks.
- The flux of gas leaving the near field may increase, including levels of tritiated hydrogen.

In mitigation of these effects:

- The availability of water may be restricted early in the post-closure phase, when the repository would not have resaturated with groundwater. This would limit the additional gas due to corrosion. pH control may however be lost above 200°C (Atkinson *et al*., 1995).
- Some of the key factors controlling the solubility of long-lived radionuclides, particularly pH and redox potential, may be little affected by a temperature rise up to 200°C (Atkinson *et al*., 1995).
- Mineralisation effects may help to overcome initial cracking due to large temperature gradients.

5.4 Prioritisation of Risks

The additional risk associated with each of the five performance indicators in the context of Wigner energy release and repository safety can be broken down into the main components of risk:

- The probability that an impact will be made on repository safety.
- The consequences for repository safety.

Both components are conventionally mapped on to risk matrices. Each aspect has been classified in qualitative terms as low, medium and high. The meanings assigned to these are as follows:

- *Probability*: Low, risk has negligible probability of arising; Medium, risk may occur; High, high probability of occurrence, based on available evidence.
- *Impact*: Low, no effect on repository safety; Medium, may affect repository safety; High, significant impact on repository safety, based on available evidence.

The risk matrices for the two scenarios studied are shown in Figures 5.1 (maximum temperature limited to 78°C) and 5.2 (maximum temperature reached assumed to be 200°C). Account has been taken of the mitigating factors noted above in drawing up these matrices.

5.4.1 Scenario with maximum temperature 78°C

All the safety indicators connected with the flux of radionuclides leaving the repository in groundwater should be little affected on this scenario. The impacts are therefore low. Taking into account the mitigating factors noted earlier, it is concluded that associated probability is also low.

The impact on repository safety in connection with gas generation could be higher, and has been assessed as medium. The associated probability is again taken to be low, based on the mitigating factors identified.

5.4.2 Scenario with a rapid rise in temperature extending above 140°C

The risk matrix changes substantially on this scenario. The probability associated with the impact of Wigner energy release on all the safety indicators is assessed as high. It seems almost certain that considerable changes will occur in and around the packages under consideration that could be on a scale sufficient to affect the safety of the whole repository.

The impact of Wigner energy release is assessed as high on three of the safety indicators:

- *Physical integrity of near-field barriers limiting the movement of short-lived radionuclides:* considerable damage could be done to near-field barriers, including the backfill, containers and grouts, permitting easier movement of short-lived radionuclides.
- *Long-term effect on groundwater pathway:* Wigner energy release could affect the flux of radionuclides leaving the repository in groundwater through increasing concentrations in solution and opening up new flowpaths. Although mitigating factors have been identified relating to both aspects (particularly below 200°C), it is concluded that together they merit assessment as a high impact on present knowledge.

- *Amount of gas leaving the near field:* although limitations on water availability could modify rapid gas generation, steam is likely to be generated from the cements present. The overall impact on repository safety could be high.

The impact on the remaining two indicators is assessed as medium:

- The consequences for *chemical control of concentrations of radionuclides in groundwater* are mixed. Some key factors (pH and redox potential) may be little affected at temperatures up to 200°C, but sorption may decrease due to mineralisation. Above 200°C, the impact may become high if control over pH is lost.
- *Short-lived changes to the groundwater pathway* (i.e. for the duration of the temperature transient) may be limited because of limitations on the availability of water in the regions of the repository affected.

5.5 Commentary

The scenarios analysed in this Section illustrate the general approach that is recommended for understanding the level of risk associated with options for packaging irradiated graphite. It is necessary to appreciate the temperature transients that can develop, the mechanism that triggers the release of Wigner energy, and the timing of this event. Reported modelling of transients at present focuses on the packages containing the graphite. In order to appreciate the effects this may have in the post-closure phase of a repository, it is necessary in addition to develop an appreciation of the knock-on temperature changes in the surrounding backfill and immediately adjacent packages. Such changes can be critical in the assessment made of the impact of Wigner energy release on repository performance. These knock-on effects have not yet been tackled in reported modelling studies (Guppy *et al.* 1999).

The assessments of temperature rise are most cost-effectively carried out by modelling, but there is a need for additional experimental back-up to confirm key aspects of the behaviour of the system, particularly for the controlled release of Wigner energy at the relatively low initiating temperatures that would exist in a repository.

The relationship between temperature transients and the risk in the post-closure period should be assessed using the framework developed here using indicators of near-field safety (see subsection 5.2). It is recommended that these indicators should be quantified as part of evolving the methodology.

The risk assessment carried out here for two options for packaging irradiated graphite wastes suggests that the additional level of risk from Wigner energy release in the packages containing 100% graphite is much higher than that containing a 50:50 mix of graphite and waste. It may be possible to engineer the emplacement of the former type of package so that the level of risk is substantially reduced (possibly by incorporating more heat sinks around the packages). Such an emplacement strategy would, however, go against the requirement to achieve passive safety (see subsection 2.6). In addition, the modelling was carried out using 'worst case graphite' from the point of view of Wigner energy release. Lower temperature excursions may be estimated if a convincing case can be made for moving away from this conservative assumption. Further mitigation may be achieved if it is not physically possible to fill the packages to the degree assumed because of the shape of the waste pieces.

In the case of the package containing a 50:50 mixture of graphite and cement, confirmation is needed that the Wigner energy will indeed be released in the controlled manner indicated by the modelling.

The scenarios assessed in this Section do not include the following issues of some potential significance:

- *Mixed wastes containing irradiated graphite*. The mixed waste in storage at Sellafield contains mostly Magnox and aluminium, besides irradiated graphite. The temperature that the metals would experience during release of Wigner energy would of course be that of the package containing the graphite. However, the non-graphite materials present (including any cementitious grout) would reduce the maximum temperature reached by providing a heat sink. There is a need to understand whether the average graphite content of each package is sufficient to lead to problems during disposal, and if not, the graphite content at which repository safety might be significantly affected.
- *Annealed graphite.* As noted in Section 3, the residual Wigner energy remaining after annealing will need to be understood. This is probably best done through a combination of the plant design process and quality checking during plant commissioning. The results can be fed into assessments of the effects of this on repository safety and waste package design. This is probably best done assuming a cautious approach, i.e. using the maximum level of residual Wigner energy and taking account of uncertainties in the modelling.
- *Wigner energy release initiated by a late event*. The analysis presented in Section 4 suggests that release of Wigner energy is likely to occur in the short term. The events that might give rise to a late release are identified in Section 4 as microbial degradation of organic wastes and corrosion of metals, particularly steels. If a late release does occur, it will follow the anticipated degradation of the near-field barriers. The first of the safety indicators identified in subsection 5.2 (concerning the physical integrity of near-field barriers) need not therefore be taken into consideration. The chemistry of long-lived radionuclides, and therefore their flux in groundwater, could be affected. Groundwater will be available to participate in corrosion, which should be mainly of steels and Zircaloy at long timescales, the reactive metals having largely corroded away. At the high hydrostatic pressure in a fully saturated repository, steam will not form until well above 100°C, again indicating that conditions could favour a burst of gas forming.

Impact on Repository Safety		Probability of Impact on Safety being Realised: Low	Medium	High
	High			
	Medium	X^5		
	Low	X^1 X^2 X^3 X^4		

Figure 5.1 Risk matrix concerning effect of Wigner energy release up to 78°C on repository safety

Note X^1 to X^5 are the risks associated with the safety indicators 1-5 respectively, as defined in subsection 5.2.

Impact on Repository Safety		Probability of Impact on Safety being Realised: Low	Medium	High
	High			X^1 X^4 X^5
	Medium			X^2 X^3
	Low			

Figure 5.2 Risk matrix concerning effect of Wigner energy release up to 200°C on repository safety

Note X^1 to X^5 are the risks associated with safety indicators 1-5 respectively, as defined in subsection 5.2.

6. KEY FACTORS IN ASSESSING PACKAGING PROPOSALS

6.1 Introduction

This Section is concerned with (a) identifying those key criteria involved in comparing options for managing graphite wastes that have emerged from the work reported in earlier Sections and (b) commenting on the four main options identified in subsection 2.5 in the context of these criteria, together with the 'do nothing' option (Environment Agency 2000). No final conclusions are drawn concerning the relative applicability of the options: this can realistically only be done in the context of detailed consideration of individual waste streams.

The criteria identified are recommended for inclusion in systematic determinations of Best Practicable Environmental Option (BPEO) for managing irradiated graphite wastes. Their applicability to the mixed graphite waste in the mixed waste silo at Sellafield depends on the scale of potential impacts to safety caused by any Wigner energy release. As noted in subsection 5.5, this has still to be determined.

BPEO is concerned with comparison of options and process optimisation. Government policy (Review of Radioactive Waste Management Policy, Cm 2919, 1995) requires "nuclear site operators to ensure that radioactive wastes are not unnecessarily created" and that the "authorisation of radioactive discharges depends not only on maximum discharges and limits of exposure but also on the optimisation of the practice". Nuclear site operators are required by the Environment Agency to optimise processes using BPEO methodologies.

The determination of BPEO is usually conducted by a team of suitably qualified and experienced personnel. The methodology employed usually falls into three stages:

1. The identification of all possible options for achieving the operation under consideration. In this case, each option should start with the treatment of the graphite wastes, and extend into the period of surface storage of the resulting solid wastes and their eventual disposal. This study is not concerned with the methods used to remove the graphite from its original location, though BPEO panels may wish to consider this stage as well. Reference is made below to *in situ* techniques for treating graphite where appropriate.
2. Screening of these options to identify those that are feasible for implementation in the short term.
3. Detailed evaluation of the short-listed options against a range of criteria defined by the panel. Scoring systems (including weighting of criteria to reflect their relative importance) are often adopted as a way of readily balancing the possibly conflicting demands of the various criteria.

The criteria identified here are those that emerge from this study. This is unlikely to constitute the full list identified by an evaluation panel: for example, issues concerned with the operational safety of plant and doses to workers lie mainly outside the scope of this study, but are likely to be included in a BPEO study.
The criteria identified from the current study are as follows:

- Feasibility of implementation of each option identified.
- Discharges to the environment - gaseous, liquid and solid.
- Decommissioning liabilities relating to plant that becomes contaminated with radioactivity during treatment of the graphite.
- Disposability and the associated issue of reworkability.
- Compliance with the objectives of regulatory requirements and National policy.
- Safety margins.
- The relative costs of the options.
- Views of stakeholders and the wider public.

These are discussed in turn in the following subsections in the context of the five main options identified in subsection 2.5:

1. Packaging (with or without cement grout and no pretreatment) → surface storage → final disposal
2. Annealing → packaging of graphite → storage → disposal
3. Incineration → packaging of residues → storage → disposal
4. Steam reforming → packaging of residues → storage → disposal
5. 'Do nothing'.

Options 2-4 also involve the formation of radioactive gases that may be discharged to the environment or trapped and converted to solid residues for ultimate disposal. The 'do nothing' option is discussed below where relevant. Further options might involve the use of incineration or steam reforming following annealing if there was a requirement to destroy the graphite after removal of the low-temperature Wigner energy: these systems are not discussed here, but many of the points made for the separate process steps will be valid.

6.2 Feasibility of Implementation

Whilst research and development work has been performed on options 1-4 above for managing irradiated graphite wastes (amounting to very substantial programmes for packaging and disposal), none have currently been completely implemented and hence completely proven. However, the research that has been reported in the public literature suggests that all the options are potentially technically feasible.

(a) Packaging (with or without cement grout and no pretreatment) → storage → final disposal

This is the currently the proposed primary route for many intermediate-level wastes. Both waste producers and disposal authorities (Nirex in the UK) have performed substantial research and development programmes into each step of the management route (packaging, storage and disposal) and substantial volumes of waste have now been packaged and are in surface stores (notably at Sellafield in the UK). Currently, little of this waste includes irradiated graphite but this is a waste stream that BNFL are now considering in the development of the Sellafield Box Encapsulation Plant.

The issues of the safe storage of graphite containing significant amounts of stored energy over a prolonged period and its subsequent disposal clearly needs to be considered, particularly as the energy may be liberated by modest temperature excursions above ambient (Guppy *et al*. 1999). The period in a repository may include in extensive period of care and maintenance while the repository is kept open. Accident scenarios (including the effect of fires) would need to be considered for both surface and below-ground storage. No evidence was found in the literature that an engineered solution has yet been determined.

(b) Annealing → packaging of graphite → storage → disposal

Whilst the possibility of annealing irradiated graphite to remove stored Wigner energy has been understood since the 1940's and crudely implemented in the operation of early graphite pile reactors, only in recent literature (Wörner *et al.* 1999 and Botzem *et al.* 1999) has the engineering of a proper facility been addressed. This work forms part of the Windscale Pile 1 decommissioning project where an annealing plant is seen as part of the processing facility for waste that will arise during the dismantling of the reactor. The research has considered the process parameters (temperature and time of heating the graphite) that need to be achieved, the method of processing, the range of sizes of graphite blocks to be annealed, the acceptability of cracks in them, and the equipment required. Whilst no plant appears to have yet been built, the process and equipment needed appears to employ current technology and to be therefore achievable. However, the process still needs to be demonstrated. Discharges to the atmosphere are considered in subsection 6.3.

The downstream activities from the annealing plant (packaging, storage and disposal) offer the same challenges as that for any ILW. With the removal of the stored energy, the possibility of a release of stored energy during storage and disposal is eradicated. However as the annealing process only removes that part of the stored energy associated with release at lower temperatures some questions remain:

- Can the gamma radiation field within the repository redistribute the energy associated with higher temperatures to lower temperatures (see subsection 2.7)?
- Does the remaining energy associated with higher temperatures increase the risk to a repository in accident conditions such as a fire?

Annealing of the mixed waste containing graphite that is stored in a silo at Sellafield would be problematic because the waste contains potentially reactive metals (Magnox and aluminium) that may corrode rapidly, liberating hydrogen (a flammable gas), when heated.

(c) Incineration → packaging of residues → storage → disposal

Incineration of irradiated graphite wastes has been considered by several authors in the literature, e.g. Wise (2000), Neighbour *et al.* (2000) and Holt (1999), but most conclude that the release of carbon-14 and resulting collective dose to the worldwide population and the emission of a greenhouse gas preclude its use unless the carbon discharge can be trapped (see (d) below) and safely disposed of.

Regarding the actual process of incineration, Holt (1999) briefly reports that incineration trials of reactor graphite have been undertaken. It was found that the graphite was not readily incinerated unless the graphite was mechanically broken up into pieces approximately 25mm in size (a process that may introduce some risk of releasing the stored energy). As with annealing, it appears likely that although no plant appears to have yet been built, the process and equipment needed employs current technology and is therefore achievable.

The downstream activities from the incineration plant (packaging of residues and secondary wastes, storage and disposal) offer the same challenges as that for any ILW and hence will not be further considered here.

(d) Steam reforming → packaging of residues → storage → disposal

Mason *et al.* (2000) report on this method of processing irradiated graphite that involves steam heating of the graphite to achieve pyrolysis. Hydrogen and carbon monoxide are typically produced by this reaction which may then be further reacted to produce water and carbonate solids. This process forms the basis for an operating plant in the USA and hence its feasibility for handling non-irradiated graphite wastes is arguably proven. However, the effect of stored energy in irradiated graphite upon the process has not been demonstrated and the graphite waste may need mechanically pretreating to achieve particles of a set size.

The process is also claimed to be suitable for treating reactor cores in-situ, by circulating carbon dioxide, nitrogen and steam or oxygen through the core. However, the feasibility of this option has not been demonstrated and the issues surrounding a safety case for such an operation (prevention of structural collapse of the core, prevention of explosion from the liberated hydrogen, effect of release of Wigner energy) would be substantial.

6.3 Discharges to the Environment

The impact of Wigner energy on discharges to the environment will occur both from any processing performed on the waste as well as on subsequent packaging, storage and disposal. Each option (excluding 'do nothing') will therefore be considered in turn.

(a) Packaging (with or without cement grout and no pretreatment) → storage → final disposal

This option offers a large discharge of solid waste to a repository but little discharge of either liquids or gases under normal conditions. These discharges will not differ from those arising from any other relatively inert ILW.

The present study indicates that should a significant release of Wigner energy occur within the repository, the event has the potential to degrade repository safety (see Section 5). The extent depends upon the composition of the waste package, and in particular the proportion of grout present, which will absorb some of the heat. This situation also need to be seen in the context of the total amount of low temperature graphite scheduled for disposal. A release of energy during packaging may also degrade packaging materials and hence reduce storage safety. These events could lead to release of gaseous and liquid products, possibly radioactive, to the environment.

(b) Annealing → packaging of graphite → storage → disposal

Considering first the annealing of the graphite, the process of heating the graphite will drive off gaseous and volatile liquids which would need to be trapped in filters or discharged in a controlled manner. One current proposal is to heat the graphite in an inert argon atmosphere and hence the discharge of carbon-14 should be limited to entrapped carbon dioxide gas and hence would be of small volume. There may be a small release of tritium as hydrogen and steam (Wörner *et al*. 2000). Reactor graphite is quite pure carbon and hence the release of other materials will be small unless the graphite waste arises from a source that has been contaminated.

The liquid components may condense within the annealing plant, leading potentially to a hazard to operating personnel and eventually a decommissioning liability. Graphite dust, which will arise from handling the graphite within the plant, will lead to similar hazards and liabilities.

The packaging, storage and disposal of the annealed graphite should offer no special risk over similarly inert ILW, provided temperatures are restricted to well below the annealing temperature (50-100°C lower). If the temperature of the graphite approaches that used in the annealing, then any remaining stored energy may start to be released, leading to increased temperatures. However such temperatures are well above those anticipated for normal storage and in a repository (though not necessarily those found in a fire) and the additional effect of any stored energy release (and consequential discharges) in such conditions may well be insignificant.

(c) Incineration → packaging of residues → storage → disposal

The incineration of graphite waste would produce a very large volume of gaseous carbon dioxide together the release of any trapped gases and volatile liquids. In addition, depending upon the temperature utilised, some contaminants in the graphite could be oxidised and particulates released. The carbon products would contain the β-emitter carbon-14, which, if released, could provide a significant collective dose to the environment (Holt, 1999). These products could be trapped through further reaction to create carbonates (Mason *et al.* 2000), but the volume of resulting waste would be several times greater than the original graphite.

Filters could be employed to trap the volatile liquids and particulates, but these would need to be discharged themselves, probably to a repository after suitable packaging. In addition, there would be a discharge of tritium. The incineration plant would become contaminated with particulates and condensed liquids from the incineration process leading potentially to a hazard to operating personnel and eventually a decommissioning liability.

The residues from the incineration would be completely free of Wigner energy. However, although the volume of residue after incineration would be much smaller than the original waste form, carbonate residues (if formed) would be much greater in volume. Mason *et al.* (2000) suggest using the carbonate residues as packing for other ILW waste, but this has not been proven or accepted by waste regulators.

(d) Steam reforming → packaging of residues → storage → disposal

The discharges for this option would be similar to those for incineration and therefore the comments above apply with the exception that the lower temperatures employed may reduce the rate of movement of gases and particulates, simplifying somewhat the management procedures involved (Mason et al. 2000).

6.4 Decommissioning Liabilities

The processing of radioactive graphite containing Wigner energy will contribute to additional decommissioning liabilities by contaminating annealing, incineration or steam reforming plant and by generating additional operational waste streams (filters, oil, cleaners, etc). Careful design of equipment can minimise these arisings. Packaging without processing may lead to no additional decommissioning liabilities if the packaging requirements remain unaltered by the presence of Wigner energy. Guppy *et al.* (1999) suggest that further dilution of graphite waste forms containing Wigner energy may be necessary however, and if so this will substantially increase the volumes required in a store and a repository and hence the decommissioning liabilities.

6.5 Disposability/Reworkability

The impact of Wigner energy during the disposal phase is limited almost entirely to the route based on direct disposal. In Section 5, it is shown that this release of energy has the potential to degrade repository safety to a significant extent (based on available evidence), depending on the composition of the waste package and the amount of low-temperature irradiated graphite sent for disposal. Detailed assessments of any additional risk to an individual from disposal of irradiated graphite would seek to determine whether the overall peak radiological risk to man would exceed the target of 10^{-6} per year for the repository and whether the dose constraint of 0.5 mSv per year to a representative member of the critical group would be exceeded in the period before control is withdrawn from the site (Baker *et al*. 1997).

The other three routes are not expected to lead to Wigner energy release in the repository. For the route based on annealing, this may not be the case if:

- The treatment step does not function as intended, so that the planned release of low temperature Wigner energy during annealing is incomplete. With annealing, there is a need to identify an upper limit to the residual low temperature Wigner energy in order to estimate the corresponding temperature rise in a repository and the significance of any effects this gives rise to.
- Radiation levels in and around the packages of irradiated graphite cause some decrease in the activation energy that needs to be supplied in order to release the Wigner energy. Although this is thought to be unlikely under repository conditions, it merits further investigation (see subsection 2.7).

Other issues concerned with disposability and reworkability are discussed in subsection 6.6.

6.6 Compliance with Policy and Regulatory Requirements

Responsibility for radioactive waste management is shared between the Government, the regulators and the waste producers. The Government decides matters of overall policy; the regulators ensure that policy is implemented; while the waste producers must manage the wastes in ways that comply with regulatory requirements.

The key current policy document is Review of Radioactive Waste Policy, Cm 2919, which appeared in 1995. Key themes in this paper with implications for regulation have already been noted in subsections 1.1 and 6.1. The list may be summarised as follows:

- Wastes should not be created unnecessarily.
- Waste management practices should be optimised. This brings with it the need to demonstrate Best Practicable Environmental Option in proposals for managing wastes.
- Wastes should be treated at an early stage so as to render them essentially passive, and therefore safe for prolonged surface storage.
- While passive safety is important for waste storage, there is also a requirement to avoid foreclosure of future options for engineered disposal.

- Reworking of packages should be practicable so as to accommodate unexpected deterioration or changes in the conditions of final management.

Considering briefly the five main management routes against these regulatory requirements:

- The possibility of energy release associated with direct disposal may not comply with the need to establish passive safety (Nuclear Safety Directorate, 2001), particularly if the waste is not grouted into the container. A claim that grouting would prevent a significant temperature rise if Wigner energy were to be released in a repository would require a safety analysis that was demonstrably sound. It would also need to be shown that any Wigner energy released during the curing of the grout would not affect significantly the quality of the resulting package. Grouting the graphite into the container may foreclose certain options, unless the package could be reworked in some way in the long term. This option has the advantage that the creation of secondary wastes during waste management is minimised.
- The route based on annealing may also lead to foreclosure of options if the treated waste is grouted into containers. It may, however, be possible to make a case that avoiding grouting may still give a package that complies with the principle of passive safety. Radioactive gases and particulates may be released during the annealing stage, but to a lesser extent than with incineration or steam reforming: this may constitute a degree of optimisation of the management route.
- Both incineration and steam reforming may lead to the creation of unnecessary wastes because essentially all of the tritium and ^{14}C will be converted to gas. This may be overcome using offgas treatments inside the plant that convert the gases to water (containing tritium) and an insoluble metal carbonate (to remove ^{14}C from the gas phase) for ultimate disposal. The volume of wastes generated may, however, rival that of the original graphite. Thus, while these processes in themselves comply with the requirement for passive safety, other aspects of the waste management system may have significant safety implications.
- A 'do nothing' strategy may be viewed favourably in terms of not foreclosing options and the avoidance of having to rework packages. This strategy does not, however, involve early waste treatment, and so the strategy may not be optimised with respect to passive safety. The balance between these issues needs therefore to be understood in order to assess a 'do nothing' option.

6.7 Safety Margins

There is no discussion of safety margins in the literature on any of the options. Determination of safety margins requires detailed assessments to be made of operational and abnormal (accident) conditions. A probabilistic approach is needed derived from a detailed analysis of the processes and events that may occur and an evaluation of the severity of each event. It is outside the scope of this study to perform such analyses and the information available from the current literature is too vague to allow the analyses to be performed.

6.8 Cost

The literature contains no information on the cost of each option and hence only general points can be made, as follows.

From a waste producer's viewpoint, there are great benefits to managing the waste stream and removing the uncertainty associated with Wigner energy as soon as possible, because future liabilities reduce stakeholder confidence and hence company value. The option to package and store the graphite waste without removing the Wigner energy may therefore look unattractive, as the liability for the waste remains and future costs may escalate if the assumptions for the storage and disposal routes are wrong.

Annealing, incineration and steam reforming should remove the issue of Wigner energy and hence some of the uncertainty. Against this, annealing, incineration and steam reforming require funds to be spent today and hence are more expensive in the short term than simply packaging the graphite.

Incineration with simple discharge of the exhaust gases (if licensed) to atmosphere may appear the cheapest option because it greatly reduces the volume of waste and costs associated with storage and disposal. However, the cost associated with poor publicity from the release of large quantities of carbon-14 (a greenhouse gas) may make this an unattractive option.

6.9 Views of Stakeholders and the Wider Public

The types of stakeholder involved in decisions concerning the management of graphite waste include:

- local communities who may be affected directly, particularly by releases of radioactivity to the atmosphere;
- the waste producers, who will be responsible for treatment and storage;
- Nirex, as the body responsible for repository development and advising waste producers;
- the regulators;
- environmental groups; and
- Government Departments.

Beyond these bodies, the wider public may also take an interest in any proposal for managing wastes from which Wigner energy may be released because the Windscale fire of 1957, which may have been due to a release of Wigner energy, remains in the public memory.

These groups may, for example, take different views (depending on the option under consideration), ranging from instinctive (or subjective) concerns about risks associated with the proposed treatment route (for example, aerial radioactive discharges) to technical and economic issues relating to fully worked out analyses (for example, where the claimed safety of the option depends on the results of modelling).
Direct disposal may raise appreciable concerns about the safety of the whole disposal facility (see Section 5). Incineration and steam reforming may lead to concerns in the local community over the possible release of gases containing radionuclides,

particularly tritium and carbon-14. The volatile isotope caesium-137 may also be a concern with graphite from Pile 1 (Wise 2000). In contrast, technical analyses may reveal that doses to the public are expected to be negligible, and that releases of radioactive species to the air conform to good practice on the site in question (Neighbour *et al.* 2000).

In this context, proposals for incineration of wastes that are not radioactive often result in considerable public debate and protest, even where it is claimed that there are no public health hazards. Such attitudes may colour the perception of the incineration option and possibly that based on steam reforming. Views may alter if it is felt that an incinerator may increase local employment, or that the proposed practice is fair in the sense that the waste was generated locally (Löfstedt 1997).

In contrast, a route based on annealing may command more widespread confidence because discharges to the atmosphere are expected to be much lower, although this issue requires further consideration (see subsection 2.5). Adequate quality control of the process will need to be demonstrated, to show that it behaves in accordance with the design intent. Issues concerning the possibility of the 'trickle down' of energy from the peak around 1200°C also need to be understood (see subsection 2.5).

The 'do nothing' option may not be favoured by the public because of the impact on society of the original fire, leading to a view that 'something should be done'. In contrast, technical and economic assessments may show certain advantages to delaying the management of graphite wastes, notably that there would be no foreclosing of future options.

Groups evaluating options may wish to consider the measures that might be taken to bridge the differences in the views of various groups. Clear communications strategies may be recommended by evaluation panels that show clearly that reasoning behind the choice of a route that results in discharges to the environment (including the comparison with competing options). Technical solutions may also be considered, for example the use of gas trapping processes to minimise aerial discharges. Technical 'fixes' may, however, lead to significant increases in cost that need to be factored into the overall optimisation.

6.10 Conclusions

Groups tasked with determining BPEO for managing a particular graphite waste stream will want to select waste management options and criteria for their evaluation that reflect the detailed circumstances of the study. Factors additional to those discussed above may include:

- The volume of the waste stream.
- The inactive materials present.
- The radionuclide composition.
- Doses to workers in managing the waste stream.
- Local circumstances at the site involved, including the overall direction of waste management plans and releases to the environment from existing plants.

The criteria identified in this study are recommended for inclusion in such evaluations, in addition to these waste, processing and site-specific issues.

When the full range of criteria are evaluated, the groups concerned may find that certain aspects require more detailed consideration. They may therefore decide to commission special studies. One example might involve the balance between the additional risk involved in not optimising passive safety in the relative short term against and the risk associated with having to rework packages if the preferred waste management route changes in the longer term. The identification of BPEO should consider complete waste management systems, taking the waste from its creation to the long-term disposal phase. The identification process should also include the 'do nothing' option, and whether the risks associated with this outweigh those associated with competing waste management systems.

The identification of BPEO should be revisited from time to time (Environment Agency, 2000) to reflect:

- the impact of technical advances in waste management systems and reworking waste packages;
- the capability of new assay techniques, for example to allow model validation to be completed;
- changes in public perception; and
- improved information and data,

to confirm and refine (as appropriate) the way forward.

7. CONCLUSIONS AND RECOMMENDATIONS

The final conclusions and recommendations to stakeholders from this study are based in the first instance on the list of outstanding issues that emerged from the literature review (see subsection 2.8). Some of these issues (numbers 8-12 in subsection 2.8) are developed in Sections 4-6, and this is reflected in the findings given below. Further work on the remaining issues (numbers 1-7) is beyond the scope of the present study, and they are recapitulated here for the sake of completeness.

(a) Open Publication

The literature review shows that there are many papers dealing with the fundamentals of the generation of Wigner energy, its measurement and release. The BNES/IAEA conference on the management of irradiated graphite wastes held in Manchester in 1999 has led to a considerable increase in the number of papers in the open literature dealing with this specific issue. **It is recommended that:**

- **further papers should appear in the open literature as the topic of managing graphite wastes matures; and**
- **independent verification and reviews of this work should be commissioned**

to help build widespread confidence in the management route(s) selected.

(b) Checking the Effectiveness of Graphite Annealing

Prior removal by annealing of the Wigner energy that might otherwise be released in a repository will require a quality checking regime for the treated graphite to confirm that the process has performed as intended. Such a regime might be based on extensive analysis of annealed graphite during plant commissioning, with temperature measurements and the time of treatment ensuring that in routine operation, the required temperature is reached throughout the graphite. **It is recommended that consideration be given to this matter.**

(c) Residual Wigner Energy after Annealing

The effectiveness of annealing in terms of the temperature rise that any residual low temperature Wigner energy may cause in a repository has yet to be demonstrated. This would form part of the justification for proposals based on annealing. **It is recommended that this aspect be addressed in such proposals.**

(d) Releases to Air

The possible use of annealing to treat graphite wastes would lead to the release of radioactive gas containing tritium (in hydrogen and water) and possibly carbon-14 and radioactive particulates. **It is recommended that these be quantified and their impacts assessed in greater detail than at present.**

(e) Management of Graphite Dust

Graphite dust will be generated during the decommissioning of graphite structures; work on the management of this waste is at an early stage of development. **It is recommended that this development work should continue.**

(f) 'Trickle-down' Effects

It is possible (though on balance unlikely) that the γ-irradiation of containers of irradiated graphite by surrounding packages in a repository could lead to the redistribution of Wigner energy levels such that more stored energy than expected could be released under repository conditions. **It is recommended that a more thorough understanding be developed of this mechanism to determine whether it has implications for management routes based on direct disposal and annealing.**

(g) System Costs of Management Options

No information exists on the comparative costs of the options identified for managing irradiated graphite. In order to facilitate the comparison of options, this should focus on the 'cradle to grave' system costs, for example, to include the costs of packaging, disposal and plant decommissioning as well as the initial annealing. **It is recommended that system costs be developed for complete waste management options.**

(h) Validation/Applicability of the Minshall model for the Release of Wigner Energy

Differential scanning calorimetry is the technique best suited to measuring that fraction of the stored Wigner energy that might be released under the temperature conditions likely to occur in a waste repository. The rates of heating graphite used with this technique are of necessity much greater than those that would occur in a repository. The validation of the Minshall model against experimental data may therefore be considered to be incomplete. The model is, however, an evolution of earlier modelling approaches, and as such could command the confidence of the scientific community. **It is recommended that the validation status of the Minshall model be reviewed with respect to the availability of experimental data and the model's acceptability in terms of the modelling approach that it embodies. From this review, a plan to improve the validation status of the model (if appropriate) should be devised.**

(i) Initiating Events for Wigner Energy Release in a Repository

An analysis of a comprehensive range of Features, Events and Processes (FEPs) that could occur in a deep repository has shown that the heat of curing of the backfill; radiolytic decay heat; and heat liberated during corrosion of metals and microbial degradation of certain organic wastes could lead to the release of Wigner energy in a deep repository. The first two events would lead to the release occurring around the time of repository closure.

It is recommended that these findings be reviewed by a suitably qualified group to confirm that no potentially suitable initiating events have been missed. It is also recommended that this analysis be complemented by one that identifies events that could lead to the release of Wigner energy during repository operations and prolonged storage.

(j) Effects of Wigner Energy Release in a Repository

A methodology has been constructed for ascertaining the degree of risk associated with scenarios in which Wigner energy is released in a repository. **It is recommended that this methodology be used for evaluating proposals for managing irradiated graphite by direct disposal or annealing, followed by disposal. It is also recommended that quantitative performance measures be developed as part of evolving this methodology.**

(k) Repository Temperature

While a repository may be able to accommodate temperature transients of more than 80ºC, a restriction appears at around 100ºC, the boiling point of water. A target upper temperature of 80ºC therefore includes a margin of safety. **It is recommended that this margin be maintained in assessing the impact of Wigner energy release, principally due to uncertainties in modelling.**

(l) Criteria for Selecting BPEO

The work carried out on this study has led to the identification of a number of potentially significant criteria in comparisons of options for managing irradiated graphite that aim to identify BPEO. The criteria relate to the environmental impacts of routes for managing graphite wastes over their entire lifetime. **It is recommended that panels conducting such assessments use these criteria**. **It is also recommended that the identification of BPEO be revisited periodically to take into account changing circumstances with respect to technology, data, public perception, etc.**

(m) Mixed Waste at Sellafield

This waste contains irradiated graphite, along with Magnox and aluminium. It is unclear whether the amounts of Wigner energy that might be liberated in a repository are potentially significant in terms of repository safety: the other materials may be capable of absorbing or dissipating much of the heat released. **It is recommended that assessments of the likely behaviour of the packaged waste in a repository with respect to Wigner energy release be carried out to determine whether it is a priority issue for this waste stream. The concentration of irradiated graphite in this waste at which safety issues arise should be determined, as should the impact of the non-graphite components of packages on heat transfer.**

REFERENCES

Agg P J, Cummings R W, Rees J H, Rodwell W R and Wikramaratna W, 1996 *Nirex gas generation and migration research: report on current status in 1994*, Nirex Science Report S/96/002.

Atkinson A, Harris A W and Hearne J A, 1995 *Hydrothermal alteration and ageing of synthetic calcium silicate hydrate gels*, Nirex Report NSS/R374.

Baker A J, Lever D A, Rees J H, Thorne M C, Tweed C J and Wikramaratna R S, 1997 *Nirex 97: an assessment of the post-closure performance of a deep repository at Sellafield. Volume 4: the gas pathway*, Nirex Science Report S/97/012.

Baker M M, Less L N and Orman S, 1966 *Uranium and water reaction. Part 1 - kinetics, product and mechanism*, Transactions of the Faraday Society, **16**, 2513.

Barlow SV, Wisbey SJ and Wood P, 1998 *Gas cooled reactor decommissioning – Packaging of waste for disposal in the United kingdom deep repository.* In Proceedings of IAEA Meeting on Technologies for Gas-cooled Reactor Decommissioning, Fuel Storage and Waste Disposal (Vienna September 1998).

Bell J C, Bridge H, Cottrell A H, Greenough G B, Reynolds W N and Simmons J H W, 1962 *Stored energy in the graphite of power-producing reactors*, Philosophical Transactions of the Royal Society of London, Series A. Mathematical and Physical Sciences, No. 1043, **254**, 361.

Billington D E and Bailey L E F, 1998 *Development and application of a methodology for identifying and characterising scenarios,* Nirex Science Report S/98/013.

Botzem W and Wörner J, 1999 *Inert annealing of irradiated graphite by inductive heating.* In Proceedings of IAEA Technical Committee Meeting on Characterisation, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Power Reactors (Manchester October 1999).

Bradford P M, Case B and Garbett K, 1978 *Aqueous corrosion of Magnox AL-80: the effects of silicate on isothermal kinetics in sodium hydroxide solutions from 90°C to 150°C*, CEGB Report RD/L/N 116/78.

Bridge H, Kelly B J and Gray B S, 1962 *Stored energy and dimensional changes in reactor graphites,* Nuclear Engineering, London: Temple Press.

BSI 7134: Section 4.2:1990 *Testing of engineering ceramics, part 4. Thermo-physical properties; Section 4.2 Method for the determination of thermal diffusivity by the laser flash (or heat pulse) method*, British Standards Institute, Milton Keynes.

Chambers A V, Williams S J and Wisbey S J, 1995 *Nirex near field research: report on current status in 1994*, Nirex Science Report S/95/011.

Cottrell A H, Lomar W M, Simmons J H W, Bell J C and Greenough G B, 1958 *The technology of the gas-cooled power reactor*, UKAEA.

Department of the Environment, 1986 *Assessment of the best practicable environmental options (BPEO) for the management of low- and intermediate-level solid radioactive wastes.*

Department of the Environment, 2000 *Multi-criteria analysis: a manual.*

Department of the Environment, Transport and the Regions and Nirex, 1999 *The 1998 United Kingdom radioactive waste inventory.*

Environment Agency, November 2000 *Guidance document for Environment Agency inspectors on ILW conditioning submissions by the nuclear industry*, Consultation Draft.

Fermi E, 1942 *Report for month ending December 15, 1942,* Physics Division, USAEC Report CP-387, University of Chicago.

Francis A J, Cather R and Crossland I G, 1997 *Development of the Nirex reference vault backfill: report on current status in 1994*, Nirex Science Report S/97/014.

Goldberg J E, Gould L J, Holtom G J, Swanton S W and Tweed C J, 1997 *Effects of elevated temperatures in the near field of a cementitious repository for radioactive waste*, Nirex Science Report S/97/013.

Guppy R M, McCarthy J and Wisbey S J, 1999 *Technical assessment of the significance of Wigner energy for disposal of graphite wastes from the Windscale Piles.* In Proceedings of IAEA Technical Committee Meeting on Characterisation, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Power Reactors (Manchester October 1999).

Harris A W, Atkinson A, Cole GB, Haworth A, Nickerson A K and Smith AC, 1997 *The performance of cementitious barriers in repositories*, Nirex Report NSS/R389.

Hennig G R and Hove J E, 1956 *Proceedings of the first international conference on the peaceful uses of atomic energy*, United Nations, **7**, 666.

Henson R W and Mounsey J A, 1961 *A linear rise calorimeter for the measurement of stored energy in irradiated graphite specimens,* Report DEG 328(W).

Henson R W and Simmons J H W, 1959 *An adiabatic rise calorimeter for measuring stored energy in irradiated graphite,* Report M/R 2564.

Holt G, 1999 *Radioactive graphite management at UK Magnox nuclear power stations.* In Proceedings of IAEA Technical Committee Meeting on Characterisation, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Power Reactors (Manchester October 1999).

House of Lords Select Committee on Science and Technology, March 1999 *Third report on the management of nuclear waste*, HL 41.

HSE and Environment Agency, March 1996 *Memorandum of understanding between the HSE and the Environment Agency on matters of mutual concern at licensed nuclear sites in England and Wales*.

Iwata T, Fujita F and Suzuki H, 1961 J. Phys. Soc. Japan, **16**, 197.

Iwata T and Suzuki H, 1963 *Proceedings of the symposium on radiation damage in solids and reactor materials*, IAEA, Vienna.

Kelly B T, 1981 *Physics of graphite*, Applied Science Publishers.

Locke J and Bailey L E F, 1998 *Overview description of the base scenario derived from FEP analysis,* Nirex Science Report S/98/011.

Löfstedt R E, 1997 *Evaluation of siting strategies: the case of two UK waste tire incinerators*, Risk: Health, Safety and Environment, **8**, 63.

Marsden B J and Davies M A, 1995 *Decommissioning graphite cores containing significant amounts of stored energy.* In Proceedings of International Conference on Nuclear Decommissioning (London 1995).

Marsden B J, Preston S D, Wickham A J and Tyson A, 1997 *Evaluation of graphite safety issues for the British Production Piles at Windscale: graphite sampling in the preparation for the dismantling of Pile 1 and further safe storage of Pile 2.* In Proceedings of Technical Committee Meeting for Gas-cooled Reactor Decommissioning, Fuel Storage and Waste Disposal (Jülich 8-10 September 1997), 213-223. Vienna: IAEA-TECDOC-10043.

Mason J B and Bradbury D, 2000 *Pyrolysis and its potential use in nuclear graphite disposal*, Nuclear Energy, **39**, 305.

Minshall P C and Wickham A J, 1999 *The description of Wigner energy and its release from Windscale Piles graphite for application to waste packaging and disposal.* In Proceedings of IAEA Technical Committee Meeting on Characterisation, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Power Reactors (Manchester October 1999).

Neighbour G B, Wickham A J and Hacker P J, 2000 *Determining the future for irradiated graphite disposal*, Nuclear Energy, **39**, 179.

Neubert T J and Lees R B, 1957 Nuclear Science and Engineering, **2**, 748.

Nightingale R E, 1962 *Nuclear graphite,* Academic Press.

Nikolaenko V A and Karpukhin V I, 1996 *Radiation annealing of defects under the effect of γ-radiation*, Journal of Nuclear Materials, **233-237**, 1067.

Nirex, 1995 *Post-closure performance assessment: near-field evolution,* Nirex Science Report S/95/009, 1995.

Nirex, 2000 *Generic phased disposal system documentation*.

Nuclear Safety Directorate (HSE), 2001 *Management of radioactive materials and radioactive waste on nuclear licensed sites.*

OECD/NEA, 1999 *Review of the Nirex methodology for scenario and conceptual model development.*

Primak W, 1955 *Kinetics of processes distributed in activation energy*, Phys Rev, **100**, 1677.

Primak W, 1956 *Fast-neutron damaging in nuclear reactors: its kinetics and the carbon atom displacement rate*, Phys. Rev, **103**, 1681.

Rappeneau J and Quetier M, 1962 *Proceedings of the fifth conference on carbon*, Pergamon Press, **1**, 317.

Rees J H, 1989 *Gas evolution and migration in repositories: current status, May 1989*, Nirex Report NSS/G112.

Reynolds W N and Goggin P R, 1960, Phil. Magazine, **5**, 1049.

Reynolds W N, Thrower P A and Sheldon B E, 1961, Nature, **189**, 824.

Reynolds W N and Thrower P A, 1963 *Radiation damage in reactor material,* I.A.E.A.

Reynolds W N, 1968 *Physical properties of graphite,* Elsevier Publishing.

Simmons J H W, 1965 *Radiation damage in graphite,* Pergamon Press.

Sykes, C, 1935, Proceedings of the Royal Society, **A 148**, 422.

Tabrizi M R, Lyon S B, Thompson G E and Ferguson J M, 1991 *The long-term corrosion of aluminium in alkaline media*, Corrosion Science, **32**, 733.

Vand V, 1943 *A theory of the irreversible electrical resistance changes of metallic films evaporated in vacuum,* Proc. Phys Soc, **A55**, 222.

Waber J T, 1952 *A review of the corrosion of uranium and its alloys*, Los Alamos Report LA-1524.

Wickham A J, Marsden B J, Sellers R M and Pilkington N J, 1998 *Graphite core stability during "care and maintenance" and "safe storage"*. In Proceeding of IAEA Meeting on Technologies for Gas-cooled Reactor Decommissioning, Fuel Storage and Waste Disposal (Vienna September 1998).

Wise M, 2000 *Management of UKAEA graphite liabilities*, Nuclear Energy, **39**, 249.

Wörner J, Botzem W and Preston S D, 2000 *Heat treatment of graphite and resulting tritium emissions,* Nuclear Energy, **39**, 353.