TECHNICAL ASSESSMENT OF THE SIGNIFICANCE OF WIGNER ENERGY FOR DISPOSAL OF GRAPHITE WASTES FROM THE WINDSCALE PILES

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Abstract. Plans to dismantle the core of the Windscale Pile 1 reactor, and to package the waste for interim storage and eventual disposal, are well advanced. UK Nirex Limited, currently responsible for identifying and developing a site primarily for disposal of the wide range of intermediate level wastes, is addressing the suitability of the waste from Windscale Pile 1, for transport to, and disposal at, a deep waste repository.

To support the decommissioning of Windscale Pile 1, information on the condition of the graphite has been sought. Despite the fire in 1957, recent sampling of regions of the core has shown that much of the graphite still contains significant residual Wigner energy. Unless it can be shown that Wigner energy will not be released at a significant rate during operations such as waste packaging or handling of the package, or after disposal, future safety cases may be undermined.

A model for the release of Wigner energy has been developed, which describes the stored energy as a set of defects with different activation energies. Initial values of stored energy are attributed to each member of the set, and the energy is released using first order decay processes. By appropriate selection of the range of activation energies and stored energies attributable to each population of defects, experimentally determined releases of stored energy as a function of temperature can be reproduced by the model.

Within the disposal environment, the packages will be subject to modest heating from external sources, including the host rocks, radioactive decay, corrosion processes and heat from curing of backfill materials in the disposal vaults. The Wigner energy release model has been used in combination with finite element thermal modelling to assess the temperature evolution of stacks of waste packages located within hypothetical disposal vaults. It has also been used to assess the response of individual waste packages exposed to fires.

This paper provides a summary of the issues raised by Wigner energy and a description of key points from the work undertaken by Nirex to model the release of residual stored energy from packaged irradiated graphite from the Windscale Piles.

1. INTRODUCTION

United Kingdom Nirex Ltd (Nirex) has been established by the nuclear industry with the task of developing a deep waste repository (DWR) for disposal of solid intermediate and low-level radioactive wastes (ILW and LLW). Nirex is pursuing conceptual designs for a repository, and is working with customers to ensure that the radioactive wastes are conditioned and packaged in a manner that will enable them to be safely and economically transported and disposed of, following a period of interim storage.

In advance of repository availability, waste producers have developed, or are developing, packaging concepts. Nirex provides advice to customers on packaging concepts to confirm that the packages will meet the requirements currently foreseen as being necessary for transport and disposal. Where appropriate, Nirex provides endorsement of the specific packaging plans by issue to the customer of a ‘Letter of Comfort’.
The Windscale Piles 1 and 2, operated between 1950 and 1957, are constructed from layers of graphite blocks keyed to each other by graphite slats. In October 1957, during a routine operation to remove the stored Wigner energy from the graphite components, Pile 1 caught fire. Both Piles were subsequently shutdown and have been under care and maintenance by UKAEA since that time. UKAEA has recently awarded a contract to dismantle the core of the Windscale Pile 1 reactor, and to package the waste in a form suitable for interim storage and compatible with Nirex plans for eventual transport and disposal.

The transport and disposal of the graphite from reactors operating at low temperatures, such as Windscale Pile 1, has raised a number of issues, including those relating to the significance of residual Wigner energy. Wigner energy constitutes a potential source of heat, which may influence processes associated with the immobilisation of radioactive materials and thus affect the development of future safety cases.

This paper provides a Nirex perspective on the issues associated with treatment, handling, storage and disposal of graphite decommissioning waste from Windscale Pile 1 containing Wigner energy. It also describes thermal modelling work undertaken by Nirex and its contractors to understand the consequences of Wigner energy release on waste package and disposal vault temperatures.

2. TECHNICAL SIGNIFICANCE OF WIGNER ENERGY TO NIREX

During storage and transport the waste packages contribute significantly to containment and isolation of their radioactive contents and protect workers and members of the public from the damaging effects of radiation which would result from inhalation, ingestion or skin contamination. After disposal the components of the waste packages also provide barriers to the migration of radionuclides and thus contribute to radionuclide isolation from man. Nirex requires that following production of a waste package, the immobilisation of radionuclides should be maintained by passive means through all phases of waste management, including so far as possible, the range of normal conditions it may encounter, and unplanned events such as accidents involving fires. Nirex also require that waste package designs are robust to uncertainties in eventual siting and design of the repository and that sound engineering practice and good science are used to provide the highest level of inherent safety of waste packages, wherever this is reasonably practicable.

Prior to publication of waste acceptance criteria for a deep repository, Nirex has implemented a strategy to facilitate early waste packaging. This is designed to minimise the risk of future reworking of packages, by providing guidance through a suite of Waste Package Specifications. The specifications provide a basis against which waste packagers can develop packaging proposals, using a range of standard waste containers, and against which Nirex can assess specific proposals and provide advice on their suitability for disposal. Specifications are provided for: Waste Package, Wasteform, Quality Assurance, Data Recording and Package Identification. The wasteform specifications have been produced to augment the waste package specifications by specifying any limits on the contents of packages, and best practice for producing a wasteform with the required chemical and physical properties. The specification covers immobilisation of radionuclides, mechanical and physical properties, chemical containment, hazardous materials, degradation processes and wasteform stability, gas generation and nuclear properties.
Materials present in the waste package that may influence immobilisation of radionuclides or the inherent safety of the waste package are therefore of interest to Nirex. In particular, a material that can provide a large source of heat could affect the immobilisation and dispersion of radioactive material. Large releases of heat by the wasteform during its production could affect product quality by generating unimmobilised material and defects in the wasteform. This could make demonstration of compliance with safety cases more difficult and put the inherent safety of the waste package into question.

Nirex has undertaken modelling and experimental studies to examine the effects of fires on radionuclide releases from waste packages and has also undertaken studies of repository performance. Although wastes that are encapsulated with a cementitious material, and which do not generate significant heat internally are not expected to suffer significant heat penetration from an external fire, if Wigner energy is released throughout the wasteform, heat penetration into the wasteform will be enhanced. This could increase releases of radioactive material in gases such as steam, or potentially affect the combustibility of the waste. Following disposal, large releases of heat could influence the effectiveness of repository barriers, for example by:

— influencing the chemistry of the backfilling or encapsulant materials, perhaps by accelerating temperature dependent degradation processes;
— generating physical defects in waste packages and backfilling materials;
— driving convective groundwater flows.

In summary, a rapid release of large quantities of Wigner energy is unlikely to be consistent with Nirex principles. In addition to raising technical issues, it also raises the issue of public and regulatory perception, although this is not covered in this technical paper.

3. WIGNER ENERGY AND MODEL DEVELOPMENT

3.1. The phenomenon

Fast-neutron irradiation of graphite in a reactor core displaces carbon atoms from their normal positions in the graphite lattice, creating a variety of types of defects through combinations of these displaced atoms. When displaced atoms return to their original state by recombining with vacancies within the lattice, a process also known as annealing, there is a release of the Wigner energy in the form of heat.

Classical theories on the accumulation and release of Wigner energy, combined with analysis of historical and recent data from Windscale Pile 1 and 2 graphite samples, has shown that the behaviour of the graphite is consistent with the existence of a spectrum of activation energies. These are presumed to be associated with the variety of defects created in the graphite by irradiation [1]. In general, as the dose received by the graphite increases, the number of defects associated with higher activation energies increases at the expense of defects with lower activation energies due to combination of smaller defects to form clusters. However, the recombination of displaced atoms and vacancies in the lattice to eliminate defects created by irradiation also occurs, the rate of this annealing process increasing with increasing temperature. As a consequence the distribution of defects “frozen” into any piece of Pile graphite may be unique, depending on its precise irradiation history, with the irradiation temperature and dose being key factors.
The release of Wigner energy from graphite in any environment subsequent to its irradiation is dependent upon the spectrum of defects created by irradiation. In general, the higher the original irradiation temperature the higher the temperature of any subsequent environment required to achieve a given rate of annealing and thus rate of Wigner energy release. Also, as explained above, the original neutron dose received by the graphite also influences the distribution of Wigner energy and thus the response of the graphite to its subsequent environment.

The total amount of stored energy in any given graphite is generally not a useful indicator of graphite behaviour. To anneal all defects in the lattice of a highly irradiated graphite within a practical period of time, and thus release all Wigner energy, temperatures of up to 2000°C would be required. At the temperatures more likely to be experienced by the graphite in practice, only a fraction of the total stored energy could be released over a relevant timescale.

Historically a number of pragmatic rules have been used to describe the relationship between irradiation temperature and subsequent release of Wigner energy. For example, one such rule states that Wigner energy can only be released at temperatures more than 50°C above the original irradiation temperature. Such ‘rules of thumb’ were convenient for a given application, since there is a strong dependence between Wigner energy release and the original irradiation temperature. However, based on the classical understanding of graphite behaviour (activation energy controlled, time and temperature dependent processes) and trends observed in experimental measurements, these approximations are not necessarily sufficiently accurate for all potential applications. In particular, a finite rate of Wigner energy release would be expected from irradiated graphite in any subsequent environment. Nevertheless, there will be a strong link between the original irradiation temperature of the graphite, and the rate of Wigner energy release observed at the temperature of subsequent environments [2].

3.2. Model development

To enable Nirex and its customers to estimate the rate of Wigner energy release from Windscale Pile 1 graphite under a wide range of conditions from wasteform production to disposal of waste packages, a variable activation energy model has been developed by BNFL based on classical theory [3,4]. The development of the model is presented by BNFL in another paper at this meeting [5] and will only be briefly considered here.

It is not currently possible to predict the spectrum of defects in irradiated graphite from first principles. The model requires selection of a range of activation energies and distribution of the stored energy between activation energy ‘bins’ in the selected range such that the model predicts a good fit to experimental measurements of Wigner energy release from a representative sample of graphite [6]. The required extent of subdivision into bins is determined by the quality of fit to the experimental results that is considered acceptable.

The experimental measurements are typically made under a specific set of conditions (e.g. a fixed heating rate and temperature range) which produce an accurately measurable heat release. A typical example of the characteristics of graphite from Windscale Pile 2 (from a graphite dowel) is shown in Figure 1 for a range of experimental conditions [7]. The calorimeter results typically show a first peak in release rate, centred at approximately 200°C
for a heating rate of 10K per minute. A second peak is often observed if experiments are continued to higher temperatures. Due to the time dependency of the release process, the temperature at which the maximum rate of release is observed will decrease with decreasing heating rates.

It is assumed that once a spectrum of activation energies and distribution of stored energy within this spectrum has been allotted to give a good fit to one set of experimental measurements, the model can be applied to a different set of conditions. For example, the conditions may be lower heating rates or lower temperatures, which may produce rates of energy release which would have been too small to measure reliably on small laboratory samples, but may be significant to large waste packages in a well insulated environment such as a disposal vault.

### 3.3. Worst-case graphite

Interactions between Nirex and its customers on the technical issues raised by Wigner energy has been facilitated through a sub-group of the Nirex Customer Technical Meeting. Samples of graphite from the Windscale Piles and measurements of the characteristics of irradiated graphite from the Windscale Piles have been made available to Nirex and BNFL by UKAEA. In addition, historical measurements on graphite from dowels that formed part of an instrumented stringer in Pile 2 have also been made available.

There is expected to be great variability in the Wigner energy release characteristics of graphite from the Windscale Pile 1, as a consequence of the range of normal irradiation temperatures and doses and compounded by the effects of the 1957 fire. Therefore there is some uncertainty about the distribution of characteristics which will be displayed by the graphite blocks. For modelling purposes, a ‘worst-case’ graphite was identified [8]. The worst-case material is defined in terms of its response to an imposed temperature, and does not contain the largest total quantity of stored energy observed within graphite from the Piles.
Therefore, it represents a case where releases of Wigner energy will be maximised at low temperatures. The Wigner energy in graphite with higher total stored energy will tend to be associated with defects that require higher activation energies for annealing. It has been argued [8] that such graphite is less susceptible to significant Wigner energy release under waste conditioning or disposal conditions than the ‘worst-case’ graphite.

In the absence of firm information to the contrary, and to allow a pessimistic assessment of the potential behaviour of packages of waste from the decommissioning of Pile 1, it has been assumed that the graphite within an individual waste package could consist entirely of ‘worst-case’ graphite. The range of activation energies selected for the worst-case graphite was divided into ten ‘bins’, each with its own inventory of Wigner energy, to give a reasonable fit to the calorimetry analysis for worst-case Pile graphite.

4. THERMAL MODELLING

A significant fraction of the decommissioning graphite from Pile 1 is expected to be suitable for packaging in the Nirex standard container known as a “4 metre box” [9]. This container and its contents is intended to satisfy the requirements for freight containers used as industrial packages Type 2 (IP-2), as given under IAEA Transport Regulations. This view is based on limited intrusive sampling of Pile 1, which indicates sufficiently low levels of contamination from the 1957 fire. The 4 metre box (4 m x 2.4 m x 2.2 m) comprises a stainless steel skin with underlying concrete radiation shielding. For the purposes of modelling, it has been assumed, consistent with proposals, that the graphite waste would be suitable for packaging in a non-encapsulated form, and therefore a heat generating binder would not be used. Consequently this stage of waste management has not been subject to thermal assessment at this stage.

The variable activation energy Wigner energy release model has been used by Nirex in combination with commercially available heat transfer codes to estimate the response of Pile graphite to expected disposal and potential fire accident conditions. The results of this work have been a factor in determining whether there may be a need for Windscale Pile 1 graphite to be annealed prior to waste packaging to release some stored energy. Such an annealing process, if required, would be intended to favourably alter the Wigner energy spectrum of the graphite, removing defects that could otherwise anneal rapidly under package production, disposal or fire accident conditions.

4.1. Disposal vault modelling

4.1.1. Vault model

For modelling purposes it was assumed that a deep waste repository vault would contain an array of 4 m boxes. An array three box lengths wide by six boxes high was specified, with any one vertical array through the vault comprising eighteen boxes, as illustrated in Figure 2. It was assumed that:

— the boxes would be stacked on top of each other with no gap and negligible thermal resistance;
FIG. 2. Waste stack arrangement for 4 m boxes in the modelled disposal vault (only one section of boxes is shown).

— there would be a horizontal gap of 0.5 m between boxes, and;
— gaps between the boxes would be filled with a backfilling material, Nirex Reference Vault Backfill, which is based on Portland cement, calcium hydroxide, limestone flour and water.

Other features, such as structural components of the vault, were also modelled [10]. Based on the assumption that the length of the vault would be much greater than its width, heat flux along the vault was assumed to be negligibly small and thus finite element analyses only considered one vertical array of 18 waste packages. It is also assumed that forced ventilation of an air space above the stack of waste packages would continue for a period after the backfilling operation.

The vaults were modelled using a finite element analysis technique with commercially available software [11]. The wasteform and shielding within the 4 m box was modelled using solid elements, and the box skin using shell elements. The vault structural features and surrounding rock were also modelled with solid elements. The calculation of Wigner energy release from the graphite was performed by a user subroutine employing the variable activation energy Wigner energy release model. The Wigner energy released was calculated on an element-by-element basis, and applied as a body heat within each element.
FIG. 3. Chequerboard arrangement of graphite and heat absorber blocks in the wasteform modelled for cases C and D: quarter section of box shown.

TABLE 1. SUMMARY OF CASES MODELLED

<table>
<thead>
<tr>
<th>Cases Modelled</th>
<th>Wasteform Designs</th>
<th>Disposal Vault Stack Arrangement</th>
</tr>
</thead>
<tbody>
<tr>
<td>A (disposal vault base case)</td>
<td>18 wasteforms per section all contain 100 % cementitious material</td>
<td>All identical packages</td>
</tr>
<tr>
<td>B</td>
<td>1 wasteform per section containing 100 % ‘worst-case’ graphite</td>
<td>Third layer from base at centre.</td>
</tr>
<tr>
<td></td>
<td>17 wasteforms per section containing 100 % cementitious material</td>
<td>Remaining waste packages.</td>
</tr>
<tr>
<td>C</td>
<td>1 wasteform per section containing 50 % cementitious material and 50 % ‘worst-case’ graphite</td>
<td>Third layer from base at centre.</td>
</tr>
<tr>
<td></td>
<td>17 wasteforms per section containing 100 % cementitious material</td>
<td>Remaining waste packages.</td>
</tr>
<tr>
<td>D</td>
<td>18 wasteforms per section all contain 50 % cementitious material and 50 % ‘worst-case’ graphite</td>
<td>All identical packages</td>
</tr>
<tr>
<td>E (fire accident base case)</td>
<td>100 % ‘worst-case’ graphite, but no Wigner energy</td>
<td>Single package only</td>
</tr>
<tr>
<td>F</td>
<td>100 % ‘worst-case’ graphite, with Wigner energy</td>
<td>Single package only</td>
</tr>
</tbody>
</table>
### TABLE 2. KEY MATERIAL PROPERTIES

<table>
<thead>
<tr>
<th>Material</th>
<th>Heat output (kW/m³)</th>
<th>Density* (kg/m³)</th>
<th>Thermal conductivity* (W/m/K)</th>
<th>Specific heat capacity (J/kg/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Graphite Wigner energy only</td>
<td>1600</td>
<td>2.1</td>
<td>Variable: 734 (0°C) to 1356 (300°C)</td>
<td></td>
</tr>
<tr>
<td>Structural concrete</td>
<td>0</td>
<td>2376</td>
<td>1.4</td>
<td>880 *</td>
</tr>
<tr>
<td>Backfill</td>
<td>Peaks at 2.7 after 4 hours (curing only)</td>
<td>1730</td>
<td>1.2</td>
<td>2100 *</td>
</tr>
<tr>
<td>Steel</td>
<td>0</td>
<td>7800</td>
<td>15</td>
<td>460 *</td>
</tr>
<tr>
<td>Air</td>
<td>0</td>
<td>1.17</td>
<td>0.03</td>
<td>1006 *</td>
</tr>
<tr>
<td>Other non-graphite wasteforms</td>
<td>0.0015*</td>
<td>1800</td>
<td>0.7</td>
<td>840 *</td>
</tr>
<tr>
<td>Host rock</td>
<td>0</td>
<td>2724</td>
<td>2.3</td>
<td>886 *</td>
</tr>
</tbody>
</table>

* assumed not to vary significantly with temperature or time.

### 4.1.2. Cases modelled

Ranges of material parameters, such as thermal conductivity and heat capacity, were selected for the 4 m box, wasteforms, and vault and disposal vault rock materials. Two sources of heat were modelled, as potential drivers for Wigner energy release. These were radiogenic heat, which was for simplicity assumed to remain constant over the short time periods considered, and the variable heat input from the curing backfilling material.

A reference ‘base case’, Case A, was modelled incorporating an array of boxes containing only non-graphite waste. In addition, a series of variants to the base case, where various key thermal parameters for the vault were varied, were also modelled, although these are not reported here. The co-disposed waste was assigned the properties of a typical cementitious encapsulant, but with small radiogenic and other heat sources to represent the effects of non-graphite waste on thermal conditions. It was assumed that this wasteform would have good thermal contact with the box shielding at all faces, except at the lid of the box.

To assess the impact of Pile graphite on waste package and vault temperatures, Case B was defined. This incorporated one box containing Pile graphite located near the centre (the centre box in layer three of the six high stack) in each vault section. This location was chosen since it is the most highly insulated location within the stack of waste packages, which will maximise the potential for Wigner energy releases. In Case B it was assumed that the Pile graphite wasteform will comprise a solid block, with a small air space surrounding all faces of the graphite block. The space was intended to simulate poor thermal contact between the waste and the box shielding, which may be expected if the waste is to be non-encapsulated.

To illustrate the factors influencing the behaviour of the wasteform, two further cases, Cases C and D, are reported here. In these cases, the Pile graphite wasteform comprised 50% graphite and 50% inert cementitious material (assigned the properties of pre-cured backfill
with no heat output) by volume. The elements of graphite and backfill were arranged in an alternating ‘chequerboard’ fashion, with the elements sized to be similar to the dimensions of the largest blocks of graphite expected to arise from Pile decommissioning. This is shown in Figure 3. In Case C only a single, centrally located, box with this wasteform type was present in each vault section. In Case D all waste packages within the vault contained this type of wasteform. Although the precise wasteform arrangement modelled may not be practicable in a real waste package, it was intended to investigate the potential impact of diluting the wasteform with a heat absorber. The cases modelled are summarised in Table 1 and the key material properties are summarised in Table 2.

4.1.3. Results

The predicted thermal evolution for Case A is shown in Figure 4. The time axis is the time after backfilling material has been cast between the boxes within the stack, which is assumed to be instantaneous. It was assumed that the temperature of all materials within the waste stack will be 30°C immediately after backfilling. At the edge of the wasteform, the first peak in temperature at 4 days is associated with the rapid and large heat input from the curing of backfilling material. The second peak at 100 days, superimposed on the decay from the heat input by the backfill, is associated with the slow heat input from radioactive decay and other heat sources within the waste. For the centre of the wasteform, the first peak is not observed due to insulation. The maximum temperature at this point is predicted to be approximately 56°C.

![FIG. 4. Temperature versus time in Case A, the base case.](image)

The estimated thermal evolution for Case B, in which one near centre box from each section of vault contains the worst-case Pile graphite, is shown in Figure 5. Initially the rate of temperature rise is lower than for the base case, as a consequence of the insulation provided by the gap between the wasteform and the box walls. However, after approximately 3 days, internal heating from a rapidly increasing rate of release of Wigner energy resulted in wasteform temperature exceeding 140°C. The analysis was terminated at this point (see Section 4.1.4).
FIG. 5. Temperature versus time in Case B.

FIG. 6. Wigner Energy content of the ten ‘bins’ within the model during the Case B analysis.
FIG. 7. Temperature versus time in Case C.

FIG. 8. Temperature versus time in Case D.
As noted above, the range of activation energies selected for the worst-case graphite is divided into ten ‘bins’, to give a reasonable fit to the calorimetry analysis for worst-case Pile graphite. The output from the subroutine for graphite at the centre of the wasteform during Case B is shown in Figure 6. It can be seen that virtually all the Wigner energy had been released from the first four bins by the time the analysis was terminated, with progressively smaller proportions of energy released from the latter six bins.

The thermal evolution of Cases C and D, in which wasteforms comprising mixed graphite and inert backfill were present, are shown in Figures 7 and 8 respectively. In Case C, where only one waste package in each section of vault contains the Pile graphite / cement material mixture, the temperature is predicted to peak after about 20 days, with the highest temperature (78°C) being at the edge of the waste. In Case D, where all waste packages contain the Pile graphite and cementitious material, the temperature of the near centre box is predicted to peak after about 30 days, with the highest temperature (95°C) being at the centre of the waste. These peaks are associated with the effects of Wigner energy release, superimposed on the heat from backfill curing.

4.1.4. Discussion

The vault backfilling material proposed for the Nirex concept produces a large initial heat input to the system, peaking at the order of 2.7 kW per m$^3$ a few hours after emplacement. This produces a rapid initial temperature rise within the backfill and the surface regions of waste packages, which eventually penetrates to the wasteform. However, the heat input from backfill decays rapidly once reaching its peak, due to completion of the curing process. Due to the size and thermal properties of the waste stack used in the model, the rate of heat loss from the centre region of the stack is very slow, as shown by the results for the base case in Figure 4. As a consequence, the heat input from the backfill takes a long time to dissipate. In addition, other smaller heat sources, from radioactive decay or from degrading wastes, which might constitute a few watts per m$^3$, will result in the accumulation of heat over long periods of time in a large disposal vault.

As the backfill cures, heat slowly penetrates into the waste packages. This outside-in heat flow gradually increases the wasteform temperature, and accelerates the Wigner energy release. Eventually the heat flow reverses, as the rate of heat release from the graphite exceeds the heat production in the disposal environment. In Case B the heat flow from the wasteform to the backfill is insufficient to prevent accumulation of heat within the wasteform and this is sufficient to result in significant feedback effects, manifested by a rapidly increasing temperature.

The maximum temperature that would arise in Case B was not estimated. The time steps operated by the model became progressively smaller, to maintain accuracy as the Wigner energy release accelerated, resulting in impractically slow analysis. In addition the accuracy of the results at higher temperature are less certain because:

- the variable activation energy model was only populated with data for rapid Wigner energy releases at temperatures less than approximately 350°C;
- the vault thermal model does not account for processes such as vapourisation of water and mass transfer, which may become significant factors at high temperatures (e.g. above the boiling point of water).
It has been concluded that based on the assumptions concerning wasteform design and graphite properties, 4m box waste packages full of Pile graphite may behave in an unstable manner in a disposal environment.

In Case C, a more progressive release of Wigner energy from the graphite occurs. Although the loading of graphite within the wasteform is half that in case B, reducing the potential heat source by 50%, the more stable response can be attributed primarily to the presence of the inert cementitious material within this wasteform. The heat transfer path is inevitably much shorter in this case compared with the all-graphite wasteform. In this chequerboard arrangement, heat released by the graphite is absorbed locally by the inert cementitious material, reducing the feedback effect. In Case B, where the graphite wasteform is represented as a single large block, heat must be transferred to the container walls and other repository components beyond.

Even in Case D, where all 18 wasteforms in each section of the disposal vault are modelled with the chequerboard arrangement, the wasteforms still remain comparatively stable, although peak temperature rises are predicted to be up to about 35°C above that predicted for the base case. As noted previously, the precise wasteform arrangement modelled in Cases C and D may not be practicable to arrange in a real waste package. However, the model does illustrate the potential impact of diluting the wasteform with an inert heat absorbing material.

### 4.2. Fire accident modelling

#### 4.2.1. Fire model

The thermal evolution of a single 4 m box waste package exposed to a 1000°C 1 hour fire was modelled. To simulate the fire, a radiant heat flux was applied to the outer surface of the box. After the period of heating, the box was allowed to cool down in ambient temperature air. This was simulated by placing a heat sink, at a constant temperature of 30°C, on the outer surface of the box. No heat inputs other than that from Wigner energy were applied.

#### 4.2.2. Cases modelled

Two cases were modelled, Case E, a base case, and Case F. In Case E the wasteform was assumed to be graphite with the same thermal properties as worst-case Pile irradiated graphite, but with no stored energy. Even though this is not realistic, since low thermal conductivity of irradiated graphite would imply the presence of irradiation damage, and therefore Wigner energy, it was necessary to generate this base case to distinguish the effects of Wigner energy release from the significant heat input from the fire. Case F was similar to Case E except the graphite was assumed to be ‘worst-case’ Pile graphite containing Wigner energy. The cases modelled are summarised in Table 1 and the key material properties are summarised in Table 2.

#### 4.2.3. Results

The thermal evolution of the wasteform at two points for Case E, the fire test base case, is shown in Figure 9. The heat from the fire penetrates the concrete shielding in the box wall
FIG. 9. Temperature versus time in Case E, the base case for the fire conditions.

FIG. 10. Temperature versus time in Case F, external fire conditions.
and raises the temperature of the edge of the wasteform to a peak of about 300°C after 8000 seconds. However, little heat penetrates to the centre of the wasteform due to the low thermal conductivity of the irradiated graphite modelled.

The thermal evolution for Case F is shown in Figure 10. In this case the temperature of the graphite at the edge of the wasteform rises much more rapidly, reaching 350°C after 1800 seconds. The graphite at the centre of the wasteform only displayed a small temperature rise during the timescale of the analysis. No results are presented beyond 1800 seconds, as they are not considered valid (see Section 4.2.4).

4.2.4. Discussion

The temperature profiles for Case E reflect the thermal lag resulting from the use of graphite with relatively low thermal conductivity. Although the heat source is removed after 3600 seconds, the temperature at the edge of the waste reaches its peak much later.

In contrast, the exposure of a box containing graphite with Wigner energy to such high temperatures indicates significant feedback. In fact, all the Wigner energy included within the model was released by graphite in reaching 350°C. Analysis of heat distribution within the graphite showed that the raised temperatures encompassed approximately 50% of the wasteform. However, the limited population of data in the variable activation energy model affected this result. As the model was only populated with data for rapid Wigner energy releases at temperatures less than approximately 350°C, therefore it would not predict behaviour at temperatures of the order of 350°C or above. It is anticipated that temperatures would continue to rise in practice, due to availability of Wigner energy associated with defects with annealing activation energies above that modelled. Release of all modelled Wigner energy would also be expected at the centre of the wasteform, given longer analysis, but this has not been explicitly demonstrated.

5. SUMMARY AND CONCLUSIONS

A model for the release of Wigner energy from irradiated graphite, developed by BNFL, has been written into a computer sub-routine and applied with a finite element modelling technique to estimate the thermal evolution of waste packages containing decommissioning graphite from Windscale Pile 1. Two scenarios have been modelled, a repository disposal vault and a fire accident.

The modelling has shown that ‘worst-case’ Pile graphite, packaged in a non-encapsulated form in a large shielded waste container, can rapidly release a significant quantity of Wigner energy when exposed to the set of disposal conditions anticipated by Nirex. Similarly, exposure of the waste package to a severe fire accident can also result in rapid release of Wigner energy. These responses could result in large temperature rises, although quantification of maximum temperature rises in the cases modelled has not been possible.

It has also been shown that, depending on the thermal properties of the wasteform, a more stable release of Wigner energy from the graphite can occur under disposal conditions. Where the graphite wasteform is represented as a single large block, heat must be transferred to the container walls and other repository components at a rate sufficient to avoid feedback.
The results of the modelling suggest that this heat transfer cannot be achieved satisfactorily. However, where the heat transfer path is reduced, by mixing the graphite blocks with blocks of inert heat absorbing material, the heat released by the graphite is absorbed locally, thus reducing the potential for feedback.

The characteristics of the wasteform as modelled are likely to be pessimistic, since graphite is unlikely to be packaged to fully utilise the volume of the box, and the graphite is likely to show a range of characteristics other than that of worst-case graphite. However, the uncertainties in the characteristics of the graphite at this stage in the decommissioning process and the uncertainty over the specific arrangement of blocks and smaller pieces of graphite in the wasteform, have not allowed potentially more realistic assumptions to be made. It should also be noted that only a limited degree of validation for the Wigner energy release model has been carried out. Although the many experimental observations of Wigner energy release give confidence in the description of graphite behaviour by the model, further validation work is likely to be required.

Nirex requires that following production of a waste package, the immobilisation of radionuclides should be maintained by passive means through all phases of waste management. Nirex also requires that waste package designs are robust to uncertainties in eventual siting and design of the repository and that sound engineering practice and good science are used to provide the highest level of inherent safety of waste packages wherever this is reasonably practicable. The potential for rapid release of Wigner energy from a waste package containing Windscale Pile graphite is not considered to be consistent with these principles, and subsequently Nirex has recommended that prior annealing should be undertaken before packaging of such graphite.

REFERENCES


