Understanding the formation and behaviour of C-14 in core graphite

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Structure of presentation

- Background
- C-14 measurements
  - Effect of operating conditions
- Leaching behaviour
- Further validation of Activation Model
- Conclusions
Understanding the formation and behaviour of C-14 in core graphite

Provide a solution or inform decisions on graphite waste management through deeper scientific understanding of the role of C-14.

Three areas of investigation:

1. C-14 specific activity of carbonaceous deposits and underlying graphite
2. C-14 leaching behaviour and the role of carbonaceous deposits
3. Activation modelling to predict C-14 distributions across graphite cores

- Journal of Environmental Protection, 2019, 10, 118-129
  The Significance of Carbon-14 in Graphite Reactor Components at End of Generation
  Martin Metcalfe, Athanasia Tzelepi

- Annals of Nuclear Energy, 2019
  The release of Carbon-14 from irradiated PGA graphite by thermal treatment in air
  M.P.Metcalf, A.Tzelepi and G.Copeland
Project scope

- Effect of specimen preparation on deposit stability
- Extend findings of preliminary study
- Activation modelling
- Extend study to AGR
- Extend Magnox study
- Leaching studies
- Reactor operating conditions
- XPS
- SEM
- XCT

The University of Manchester
Project scope

Extend findings of preliminary study

Extend study to AGR

Activation modelling

Use of ‘enclosed’ and ‘vented’ installed set samples.
- Is it the coolant or the graphite that has the highest contribution in the C-14 content of deposits?

Leaching studies

Previous PhD student seconded to NNL: Very high C-14 release rate at the start of leaching.
- Is this initial mobility related to the deposits?
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Treatment of samples

Thermal oxidation in air of graphite trepanned from the fuel channel wall of a Magnox reactor moderator brick

- Low temperature oxidation to quantify deposit
- High temperature oxidation
- Pyrolysis and C-14 analysis

Diagram showing the process:

1. 120 mm x 6 mm disc
2. A
3. B
4. High temperature oxidation
5. High temperature oxidation
6. Pyrolysis and C-14 analysis
7. Air in
8. Air out
9. Bubblers
10. TGA

Thermal oxidation in air of graphite trepanned from the fuel channel wall of a Magnox reactor moderator brick.
Schematic showing octagon-square brick arrangement at Oldbury

Installed set carrier
Dismantling installed set carriers:
Vented sample

Dismantling installed set carriers:
Enclosed sample
Previous findings (Fuel Channel Wall – High deposit concentration)

- Carbonaceous deposits from 2 different Magnox reactor cores have C-14 specific activities with factors ~90 and ~40 higher, depending upon reactor, than those in the underlying graphite.
- Underlying C-14 activity in the graphite broadly similar.
C-14 activities (Bq/g) in deposits and underlying graphite
C-14 activities (Bq/g) in deposits and underlying graphite
C-14 activities (Bq/g) in underlying graphite

![Graph showing C14 Specific Activity (Bq/g) vs Thermal Mass Loss Increment (%)]
The significance of Carbon 14 in reactor graphite components at end of generation

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• Leaching behaviour

• Further validation of Activation Model

• Conclusions
NNL experimental programme

Leaching tests on graphite with and without carbonaceous deposits

- Investigate possible link between C-14 in deposits and mobile C-14 fraction during leaching

B Hagos, PhD thesis “Microstructural and Chemical Behaviour of Irradiated Graphite Waste under Repository Conditions” (2013), University of Manchester
The significance of Carbon 14 in reactor graphite components at end of generation

Release of C-14 by aqueous leaching

[Graph showing cumulative total activity (Bq/g) over time (days) for different dose rates (dpa).

[B Hagos, PhD thesis “Microstructural and Chemical Behaviour of Irradiated Graphite Waste under Repository Conditions” (2013), University of Manchester]
Findings - Leaching

- Even though a similar trend of the leaching rate was observed for the samples without deposits, the C-14 activity leached after the first 5-10 days is significantly lower.

- The mobile fraction of C-14 is not related to the carbonaceous deposits but the longer term C-14 activity release is reduced by ~30%.
The significance of Carbon 14 in reactor graphite components at end of generation

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Objectives

- **Effect of specimen preparation on deposit stability**
- **Extend findings of preliminary study**
- **Activation modelling**

**Objectives:**

- Assess the variability of impurities in the as-manufactured graphite
- Assess the differences in impurity levels between Wylfa and Oldbury core graphite
- Refine the NNL activation model for predictions of C-14 levels in UK graphite cores
NNL activation model

• The initial graphite composition was the same for each sample.

• A number of WIMS, TRAIL, FISPIN runs were generated in order to model the effect of graphite weight loss over the total period of irradiation of the graphite.

• For each sample, on completion of each refuelling cycle, the irradiated graphite composition was used as the input to the next FISPIN case until the final irradiation was achieved.

• NNL measurements on Oldbury R2 core graphite from the following positions:
  • Fuel Channel G85B5 Layers 4, 5 and 6
  • Fuel Channel J81B5 Layer 6
  • Interstitial Channel P82 Layers 3 and 4

[Mills et al, ENC2012, European Nuclear Conference, Manchester, UK (December 2012)]
Comparison between NNL 3D model and C-14 measurements

- Manufacturers’ data lists 10 wppm N but model calibrations against C-14 measurements indicate large variations across fleet (0-10 wppm) [Mills et al, ENC2012, European Nuclear Conference Manchester, UK (December 2012)]
Conclusions

• Measurements on
  • Interstitial channel samples
  • Enclosed and Vented Installed Sets
  • *No difference in the C-14 content of the deposits or the underlying graphite were found even though the deposit levels on these samples are very low*
  • *Indication that contribution of the coolant/air in the C-14 activity is very low.*

• Leaching experiments on samples after deposits have been removed
  • *Similar trend with plateau*
  • *Longer term C-14 release is lower than samples with deposits.*

• Validation of the Activation Model
  • *Nitrogen impurity: up to 10 ppm.*
Thank you for your attention

Any questions?
Background

Results of C-14 mass balance for an operating Magnox reactor (Metcalfe and Mills)

Measurement and activation modelling indicated an average N impurity of 10 wppm and a average C-14 activity in the graphite of $9.55 \times 10^4$ Bq g$^{-1}$.

<table>
<thead>
<tr>
<th>Mass Balance Model</th>
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<tbody>
<tr>
<td>Total C-14 activity released from core</td>
<td>1604 GBq</td>
</tr>
<tr>
<td>Total C-14 activity generated within gas coolant</td>
<td>257-281 GBq</td>
</tr>
<tr>
<td>Total C-14 production over selected period</td>
<td>~1870 GBq</td>
</tr>
</tbody>
</table>

<table>
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<tr>
<th>Site measurements reported to regulator</th>
<th></th>
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</thead>
<tbody>
<tr>
<td>Total C-14 released over selected period</td>
<td>1790 GBq</td>
</tr>
</tbody>
</table>

Agreement to within 5% between model and measurement.
Analysis of TGA data

- Slow oxidation at 450°C in air
- Monitor mass loss with time
- Fit oxidation model to data that accounts for separate deposit and graphite oxidation paths
- Deposits more reactive (red curve) than
- Underlying graphite (green curve)

- Determine deposit concentration
- Use second stage fast oxidation to evaluate C-14 specific activity in underlying graphite
- Total activity released in slow oxidation can then be partitioned between deposit and graphite
NNL experimental programme

Thermal oxidation in air of graphite trepanned from the fuel channel wall of a Magnox reactor moderator brick

- To quantify surface carbonaceous deposit concentrations
- Measure C-14 activities in deposits and the underlying graphite
- Characterise C-14 distribution within graphite