C-14 enrichment of surface deposits on Oldbury reactor core graphite

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Graphite was used as a moderator and reflector material in the first generation of UK Magnox nuclear power reactors. As all of these reactors are now shut down there is a need to examine the concentration and distribution of long lived radioisotopes, such as C-14, to aid in understanding their behaviour in a geological disposal facility. A selection of irradiated graphite samples from Oldbury reactor one were examined where it was observed that there is a distinct deposit on the exposed channel wall face surfaces that is relatively enriched with C-14, which can be distinguished both visually and using Raman spectroscopy. Although the majority of C-14 may be associated with the graphite structure, the presence of a C-14 rich surface layer needs to be understood because of the possibility that it more readily releases C-14 after closure of a geological disposal facility.

Introduction

The decommissioning of the first generation, Magnox, nuclear power reactors in the UK will lead to approximately 57,000 tonnes of irradiated graphite waste that requires disposal [1]. The current UK baseline for this waste is classification as intermediate level waste (ILW) and disposal in a geological disposal facility (GDF). This classification is due to the presence of long lived radioisotopes, including a major proportion of C-14 [2]. This radionuclide can be produced from multiple precursors and is significant for safety assessments of a GDF in the UK due to its long half-life (5730 years) and its potential to form gaseous species that may be released post closure of a GDF [3].

$^{14}N \xrightarrow{n,p} {}^{14}C$

$^{13}C \xrightarrow{n,\gamma} {}^{14}C$

$^{17}O \xrightarrow{n,\alpha} {}^{14}C$

Formation mechanisms for C-14 in irradiated graphite

Raman Spectroscopy

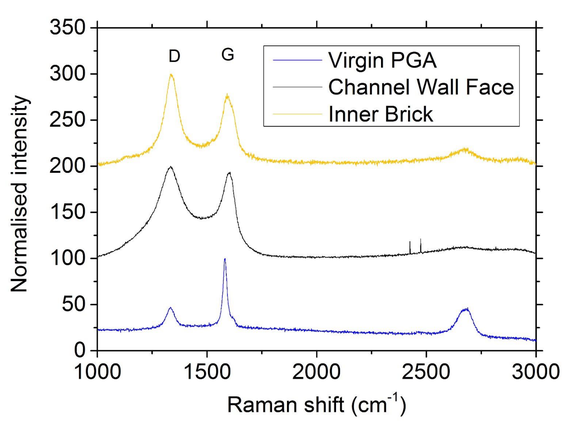

Figure 3. Raman spectra showing the broadening of the D and G peaks in irradiated graphite and the increased D broadening in channel wall face surfaces.

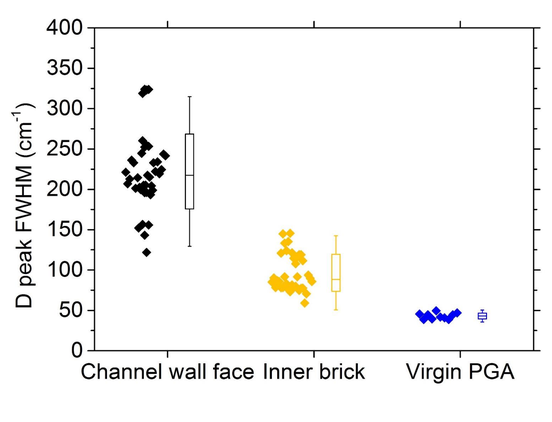

Figure 4. D peak FWHM from irradiated (channel wall face and inner brick) and virgin PGA graphite showing a method for distinguishing the three sample types.

Scanning Electron Microscopy (SEM)

Scanning electron microscopy was used to determine if there were any differences between virgin PGA graphite, inner brick and channel wall face irradiated PGA graphite, Figure 1.

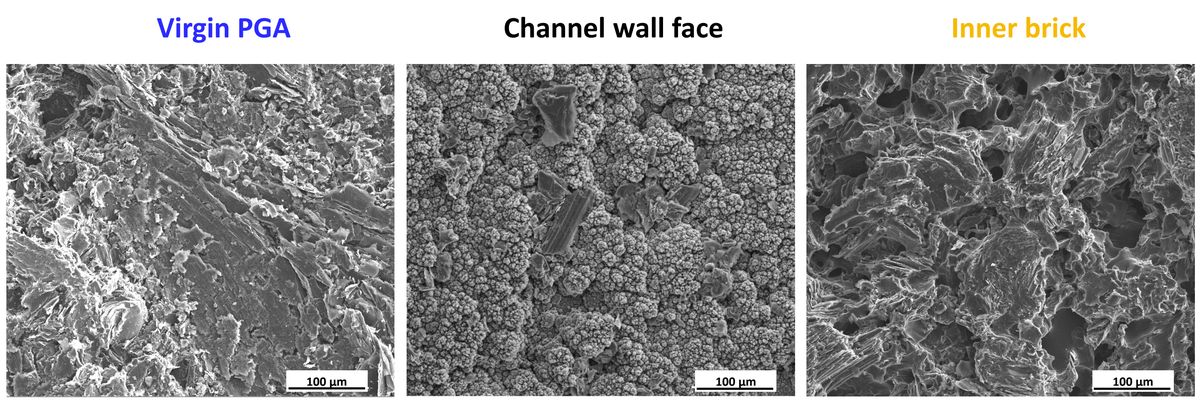

Figure 1. SEM images showing differences in the surface structure of virgin PGA graphite, channel wall face irradiated graphite and inner brick irradiated graphite.

Secondary Ion Mass Spectrometry (SIMS)

The methodology used for these determinations ensured that possible mass interferences between C-14 species and oxygen and nitrogen bearing species were eliminated from the analysis. This work indicates that the deposit found on exposed channel wall face samples has a relative C-14 enrichment compared to the underlying graphite, Figure 4. Inner brick C-14 concentrations were below the limits of detection for the instrument.

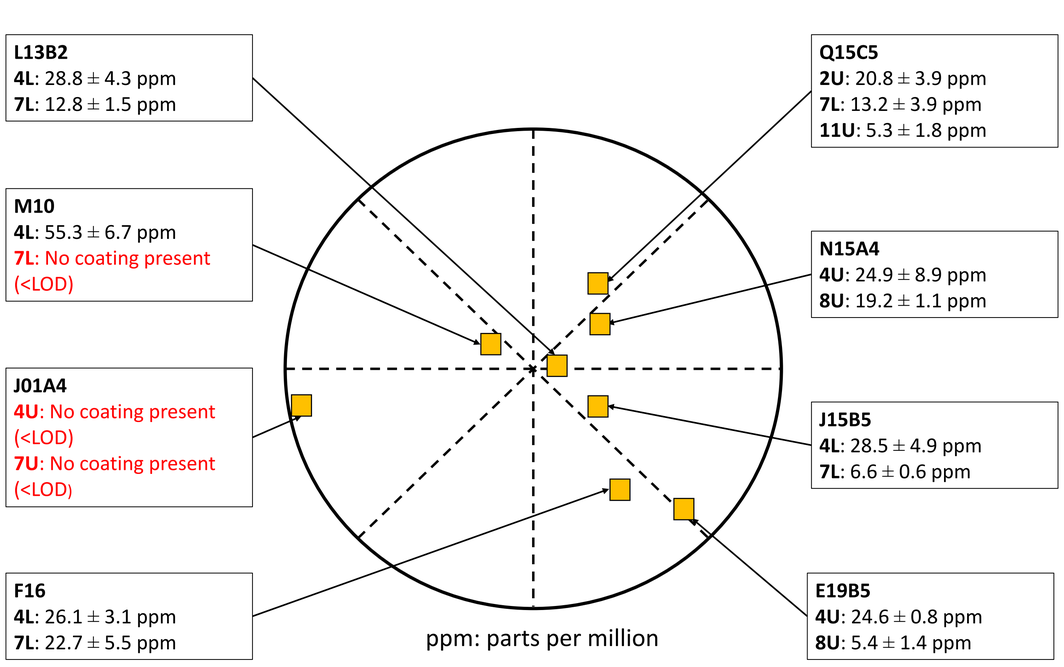

Figure 2. Results from SIMS showing the C-14 concentrations of channel wall face samples and their location within the reactor.

Thermal Oxidation / Liquid Scintillation Counting

Four irradiated graphite samples were examined to determine the concentration of C-14 in the deposit and remaining graphite. This was achieved was achieved by sequential thermal oxidation in air at 450 °C and 600 °C and capturing any gas produced in a series of bubbler solutions that were analysed using LSC, Figure 5.

It was observed that the surface deposit was relatively enriched with C-14, Table 1, with samples originating lower in the reactor exhibiting a higher concentration of C-14.

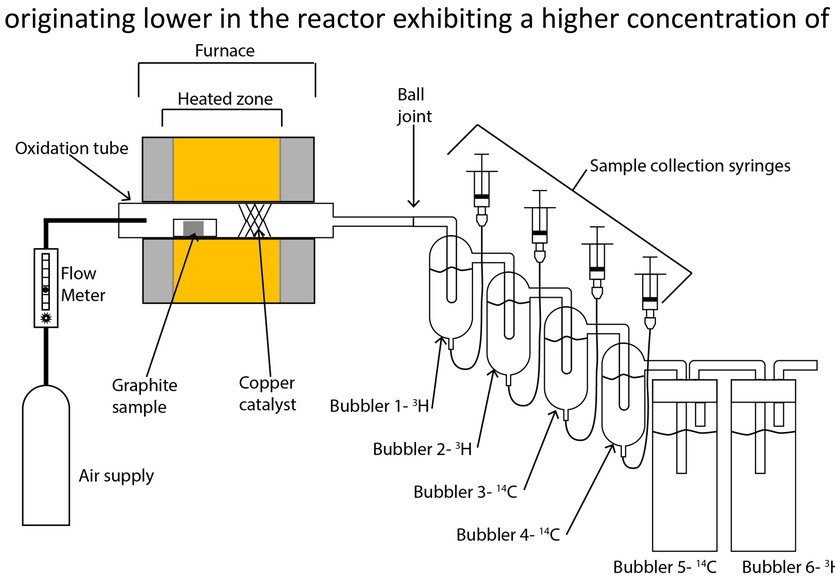

Figure 5. Labelled schematic of experimental apparatus used in the thermal oxidation of irradiated graphite.

Sample	Height in reactor (m)	C-14 concentration SIMS (ppm)	C-14 concentration LSC (ppm)
Q15C5 2U Slice 1	1.2	20.8 ± 3.9	20.9
Q15C5 2U Slice 2	1.2	4.1 ± 3.4	2.8
Q15C5 7L Slice 1	5.1	13.2 ± 3.9	9.9
Q15C5 11U Slice 1	8.5	5.3 ± 1.8	3.5

Table 1. Comparison of C-14 concentration determined by SIMS and LSC on channel wall face surfaces.

Conclusions

The research performed on irradiated graphite has led to the following main observations:

- Samples exposed to channel wall face (usually) have a pronounced carbonaceous deposit present that can be distinguished using SEM and Raman spectroscopy.
- Inner brick samples do not have such a deposit but have microstructural changes present associated with a lifetime in a nuclear reactor.
- SIMS and LSC analysis highlights a relative enrichment in C-14 on the channel wall face deposits.
- This enrichment appears to be influenced by location within the reactor but not with lifetime neutron dose.

* This work was performed at the University of Bristol, the lead author is now at RWM.

Acknowledgements

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