CARBON-14 CONTENT OF CARBONACEOUS DEPOSITS IN OLDBURY CORE GRAPHITE

M.P. METCALFE†, A TZELEPI† and J GILL*

†National Nuclear Laboratory, Reactor Operations Support, United Kingdom

***University of Central Lancashire, Preston, United Kingdom**

Abstract

Graphite specimens taken from the surface of Magnox reactor core components have been studied by differential thermal oxidation and counting of off-gases to quantify the contribution or otherwise of carbonaceous deposits to the C-14 inventory of the core graphite. While present within the open porosity of the graphite, such deposits formed from polymerization reactions in the CO_2/CO atmosphere of the reactor and from the decomposition of methane are concentrated predominantly on the outer surfaces of components. An improved understanding of their C-14 content could influence handling of material during decommissioning and could influence treatment options.

1. INTRODUCTION

This report provides the results and interpretation of the thermal gravimetric analysis (TGA) of Oldbury reactor graphite and the distribution of C-14 activities present in irradiated graphite and carbonaceous deposits. The rationale and implications of the work conducted in this project are presented below.

1.1. Background

The decommissioning of nuclear power plants around the world will produce a major waste stream of irradiated graphite. Graphite has been used extensively as a reactor moderator and reflector material that becomes irradiated and contaminated over time. In the coming years approximately 250,000 tonnes of irradiated graphite will require management making this a significant waste management issue worldwide [1].

The majority of irradiated graphite waste in the UK is classified as Intermediate Level Waste (ILW). ILW is specified as material whose activity exceeds 12GBq per tonne of beta/gamma activity or 4GBq per tonne of alpha activity and where temperature change does not have to be taken into consideration [2]. Although the activity of irradiated graphite is below these limits, it is still deemed ILW. This is due to the fact that it cannot be accepted at the Low Level Waste Repository due to the restrictive C-14 regulation at the site, as well as the volume restriction.

In addition to volume, a final waste management scheme has to take account of several important factors including stored energy, the activation of impurities and isotopic inventory. One isotope of key importance is Carbon-14 (C-14) which is a long lived isotope with a half-life of 5730 years and can be easily released into the biosphere.

C-14 is unavoidably formed in nuclear reactors due to the neutron activation principally of Nitrogen-14 (N-14), Carbon-13 (C-13) and Oxygen-17 (O-17) (Table 1). In most reactors, the rate of production of C-14 from these three main sources N-14: C-13: O-17 is in the ratio 305:33:1. Depending upon the N-14 impurity in the graphite, this C-14 precursor can potentially be a major contributor to the C-14

inventory. The amount of N-14 impurity will vary from batch to batch of material and will differ between different types of graphite used in different reactors around the UK. Another potential route for C-14 production is activation of precursors in the reactor coolant. Reactor coolants in UK reactors comprise principally carbon dioxide with small quantities of carbon monoxide, injected gases (hydrogen and methane) on some plant and hydrogenous impurities [3].

TABLE 1 MAIN ROUTES OF C-14 PRODUCTION IN NUCLEAR REACTORS [4]

Reaction	Capture cross section (barn)	Abundance of isotope in natural element (%)
$^{14}N(n,p)^{14}C$	1.86	99.63 ^{14}N/Nitrogen
$^{13}C(n,\gamma)^{14}C$	0.00137	1.11 ^{13}C/Carbon
$^{17}O(n,\alpha)^{14}C$	0.235	0.037 ^{17}O/Oxygen

Varying amounts of carbonaceous deposits have been identified on irradiated graphite samples removed from reactor cores [3]. If these deposits are rich in C-14, treatment of the waste graphite by oxidation could reduce the C-14 inventory of the remaining graphite. There is currently no deep geological repository available in the UK and the Low Level Waste (LLW) repository at Drigg has very strict guidelines regarding C-14 level [5]. The determination of the distribution of C-14 within the matrix of irradiated graphite and in carbonaceous deposits could be a key factor in the determination of possible treatments and eventual storage/disposal routes of the waste graphite.

1.2. Project information

The experimental work was carried out by an EPSRC Industrial CASE award PhD student (third author) from the University of Central Lancashire. The placement of the student in the Preston Laboratory was funded by NNL.

The study involved the use of TGA to oxidise graphite samples whilst collecting the carbon dioxide produced. The experiments described in this report comprised three procedures. A graphite disc was first split into three pieces. Piece 1 samples were analysed for total C-14 content by pyrolysis. Piece 2 samples underwent TGA to characterise deposit concentrations and underlying graphite and deposit oxidation behaviour. From this data, the time required for removing the deposits by oxidation could be determined. The same experimental parameters were used for the TGA of Piece 3 samples, this time treating the material for the time necessary to remove the deposit determined from Piece 2 samples. A second period of oxidation on Piece 3 was used to determine the C-14 content of the underlying graphite. The residue of Piece 3 then had its total C-14 content measured by pyrolysis to enable the C-14 mass balance on Piece 3 to be compared with the total C-14 measured on Piece 1.

2. METHODOLOGY

This section of the report provides experimental methodology i.e. designs and techniques used as well as information on the samples used during the study. Established NNL analytical procedures were used.

2.1. Sample Receipt

Discs sectioned from six cores trepanned from an Oldbury Magnox reactor fuel channel were used in these experiments; they were stored at Sellafield before being delivered to Springfields. Discs from the fuel channel end of the trepanned cores were chosen because they have a higher concentration of carbonaceous deposit. These discs, having dimensions of ~ ø12 x 6 mm, were dissected into three pieces by cutting the disc in half and then cutting one piece in half again (Figure 1).

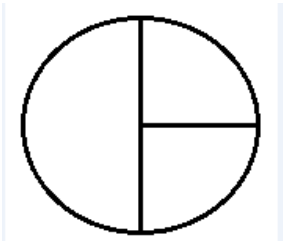

FIG. 1. Diagram showing approximate dissection of samples

The sample preparation was undertaken prior to delivery to Springfields. Upon arrival at Springfields, Piece 1 samples (Table 2) were transferred to the Measurement and Analysis Team. The selected cores were from three different fuel channels with unique reactor identifiers J17A4, J17C5 and Q15C5. Using the sample Q15C5 _2/1 (3U) as an example, the 2/1 identifies the sequential core number when the channel was trepanned (core 2) and the radial position in the core (each core is sectioned into three 6mm long discs, so disc 1 is the first at the channel wall). The identifier in brackets (3U in this example) denotes whether the core is from the Upper or Lower position (Upper in this case) and from which graphite layer counting from the bottom of the core (3 in this example).

TABLE 2 SAMPLE REFERENCES AND WEIGHTS

NNL Identity	Piece 1 Weight (g)	Piece 2 Weight (g)	Piece 3 Weight (g)	Total Weight (g)	Axial Height in core (m)
J17A4_13/1 (9U)	0.414	0.203	0.195	0.812	6.85
J17A4_8/1 (6U)	0.256	0.610	0.285	1.151	4.62
J17C5_8/1 (6U)	0.225	0.475	0.240	0.940	4.62
Q15C5_2/1 (3U)	0.206	0.399	0.220	0.825	1.98
Q15C5_3/1 (4L)	0.174	0.397	0.209	0.780	2.66
Q15C5_4/1 (4U)	0.342	0.676	0.371	1.389	2.95
Q15C5_8/1 (6U)	0.712	0.397	0.383	1.492	4.62

2.2. Complete Carbon-14 content (Piece 1)

A full C-14 analysis was carried out by the Measurement and Analysis Team using the Piece 1 samples (Table 2). This was achieved by complete pyrolysis of the graphite samples in a furnace at 800°C in an oxygen rich atmosphere in the presence of a platinum alumina catalyst. Any C-14 present

was converted to CO_2 and captured in a series of gas-bubblers containing Carbosorb. An aliquot of the trap liquor was mixed with scintillation cocktail and C-14 was determined by UKAS accredited liquid scintillation counting.

2.3. Thermal gravimetric analysis (Piece 2)

Thermal gravimetric analysis was carried out on the Piece 2 samples to determine the time required for removing the deposits by oxidation. A Netzsch 449C STA TGA with an air flow of 10 ml/min was employed for all experiments. The sample was held for 2 hours at 200°C to ensure any moisture had been removed, then ramped up to 450°C and held at this temperature for 50 hours. The furnace was allowed to cool down to 200°C and held for 2 hours at this temperature before returning to ambient temperature. The temperature program used was a standard NNL procedure. Effluent gases were passed through four gas bubblers in series to capture the Tritium (H-3) and C-14 produced (Figure 2). Dreschel bottles 1 and 2 each contained 100 ml 0.1M nitric acid for the collection of H-3. Dreschel bottles 3 and 4 each contained 100 ml of Carbosorb for the collection of C-14. Two Dreschel bottles were used for each isotope in case the scrubber solution in the first bottle became saturated. The safety case for operating the experiments required H-3 to be collected and disposed of in an appropriate manner (the solutions were not analysed). While the gases produced during the Piece 2 TGA were passed through gas bubblers, the solutions were not analysed as this information was obtained from the Piece 3 analysis.

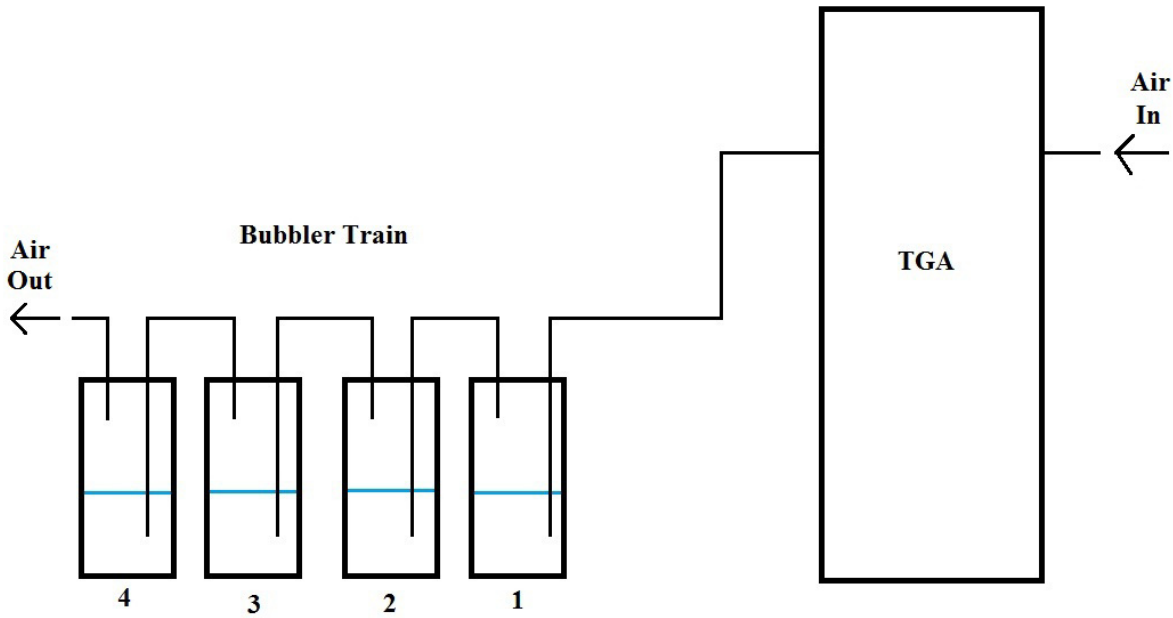

FIG. 2. Schematic of experimental set-up

2.4. Piece 3 analysis

The analysis of the Piece 3 samples involved several stages. Firstly the same TGA experimental parameters as Piece 2 samples were used with the exception that oxidation time at 450°C was based upon the estimated time for the removal of the deposit (which was determined from the Piece 2 samples). The cooling period provided adequate time to completely flush out the system before the collection of liquor from the bottles. The bottles were replaced with fresh liquor before the oxidation of the graphite sample continued for a further ~100 hours to produce a 1-2% weight loss in the specimen (the actual duration was determined from the oxidation data generated from Piece 2).

The Carbosorb samples allocated to deposit and 1-2% underlying graphite as well as the remaining solid graphite samples was sent to the Measurement and Analysis Team for C-14 analysis. An aliquot of each of the Carbosorb gas bubbler samples was taken and the C-14 activity was obtained by liquid scintillation counting. The C-14 content of the residual solid graphite was evaluated by fully combusting the samples and trapping the CO2 produced in Carbosorb, for measurement by liquid scintillation counting, as with the Piece 1 samples.

One Piece 3 sample (J17C5_8/1 (6U) in Table 2) was progressed as above but included an extended oxidation following the removal of the deposit. This experiment was included to measure the rate of C-14 released with increasing mass loss. The sample was taken to ~4% weight loss with the liquor bottles being changed at four ~1% increments. The C-14 content was determined as described above.

2.5. Quality assurance

In order to ensure the quality of the data produced a number of quality assurance steps were put in place. The performance of the TGA apparatus was tested at the start of the project and half way through the experimental proceedings using a calcium oxalate standard. The drift in the apparatus over the period of oxidation was measured from an empty crucible 'blank' run, performed after every two sample runs following the same sample temperature program. The temperature in the laboratory was logged during the experiments in the event that results would have to be corrected for lab temperature. Subsequently, it was decided that the application of this correction did not improve the data traces. An excess amount of Carbosorb was used in the gas bubblers to ensure all CO_2 produced from the experiments was collected.

3. RESULTS

This section provides the results and interpretation of the TGA of Oldbury reactor graphite and the partition of C-14 activities between irradiated graphite and carbonaceous deposits.

3.1. Complete C-14 content (Piece 1)

C-14 was positively identified in all of the Piece 1 graphite samples. Activity of C-14 has been quoted in Bq/g (Table 3 and Figure 3) and it ranges between 1.50E+05 Bq/g and 7.29E+05 Bq/g. As expected, the activity of the Q15C5 samples decreases as their axial height in the core increases. The lower position samples from graphite layer 4 shows a factor 2 higher activity compared to the upper position sample. As will be seen in the TGA analysis below, there is a factor 2 difference in deposit concentration for these two samples which may explain the difference in activity. It is unlikely that C-14 precursor impurity levels would vary to this extent within a brick. Sample J17A4_13/1 (9U) has been removed from the highest axial height in the core and exhibits the lowest amount of activity at 1.50E+05 Bq/g.

3.2. Piece 2 thermal gravimetric analysis

Piece 2 samples were subjected to TGA to determine the time required for removing the deposits by oxidation (Figure 4). Sample J17A4_8/1 exhibited an unusual profile and so was rejected from any further analysis. The other profiles show that the deposit present has a much faster oxidation rate than the underlying graphite, as expected; all samples lost ~1-3%. The time required for the removal of deposits ranged from 13-28 hrs (Table 4).

TABLE 1 - PIECE 1 C-14 ANALYSIS RESULTS

NNL Identity	Piece 1 weight (g)	C-14 Activity (Bq/g)	Axial Height in core (m)
J17A4_13/1 (9U)	0.414	1.50E+05	6.85
J17A4_8/1 (6U)	0.256	2.98E+05	4.62
J17C5_8/1 (6U)	0.225	3.50E+05	4.62
Q15C5_2/1 (3U)	0.206	7.29E+05	1.98
Q15C5_3/1 (4L)	0.174	5.78E+05	2.66
Q15C5_4/1 (4U)	0.342	2.46E+05	2.95
Q15C5_8/1 (6U)	0.712	1.59E+05	4.62

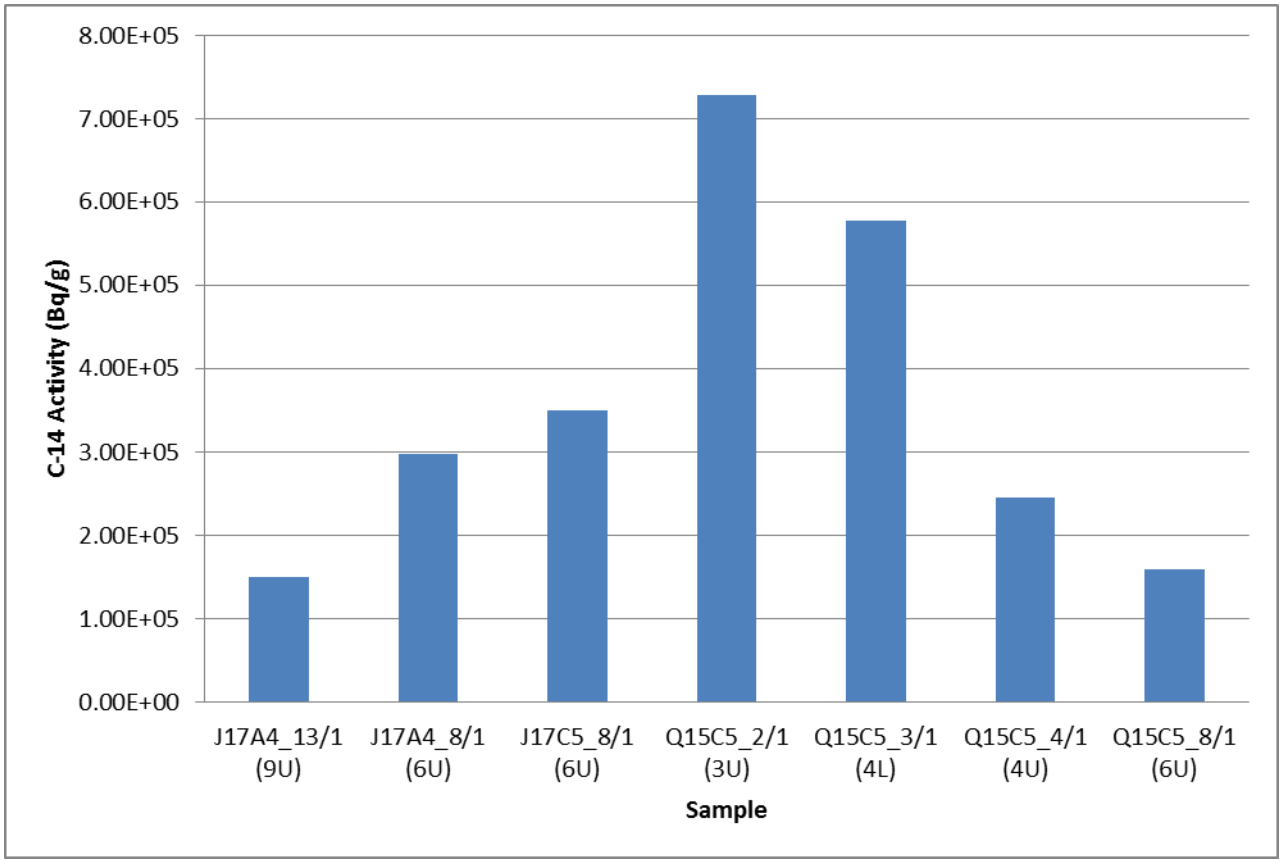

FIG.3. C-14 Activity for Piece 1 Samples

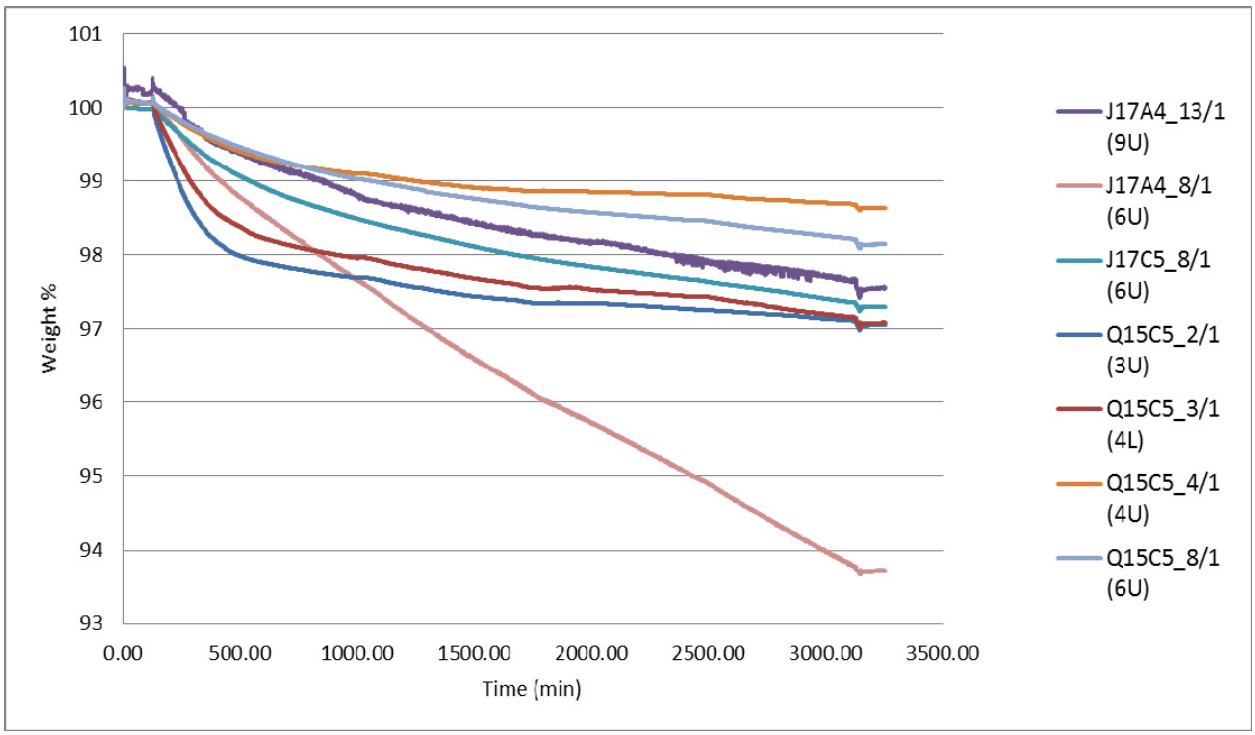

FIG.4. Piece 2 TGA Profiles

TABLE 2 - DEPOSIT OXIDATION TIMES

NNL Identity	Deposit oxidation time (hours)
J17A4_13/1 (9U)	28
J17C5_8/1 (6U)	28
Q15C5_2/1 (3U)	13
Q15C5_3/1 (4L)	16
Q15C5_4/1 (4U)	26
Q15C5_8/1 (6U)	28

3.3. Analysis of oxidation data

The oxidation kinetics of both graphite and the associated carbonaceous deposit in air at 1 atmosphere can be summarised by simple equations, which form the basis of an analytical model for TGA outputs. Using this model, a Mathematica program was used to calculate the deposit concentration (µg deposit per g of graphite) , deposit reactivity (g of deposit per g of sample per hour) and graphite reactivity (µg of graphite per g of sample per hour) (Table 5).

Deposit concentrations range from 9990-24699 µg/g. The Q15C5 samples exhibit a trend of deposit concentration decreasing with increasing axial height up the core, as expected. This trend in deposit concentration is the same shown by historical Oldbury graphite samples. Furthermore, the results from

Piece 1 C-14 activity portray the same trend: as axial height increases the C-14 activity decreases. Samples J17A4_13/1 (9U) and J17C5_8/1 (6U) have very similar deposit concentrations. Although the former sample is from the highest location in the core, the samples originate from different channels.

TABLE 3 - PIECE 2 CONCENTRATION AND REACTIVITY DATA

NNL Identity	Deposit concentration ($\mu g\ g^{-1}$)	Deposit reactivity ($g\ g^{-1}\ h^{-1}$)	Graphite reactivity ($\mu g\ g^{-1}\ h^{-1}$)	Axial Height in core (m)
J17A4_13/1 (9U)	13224	0.167	300.0	6.85
J17C5_8/1 (6U)	14350	0.183	290.3	4.62
Q15C5_2/1 (3U)	24699	0.361	129.4	1.98
Q15C5_3/1 (4L)	20759	0.297	191.8	2.66
Q15C5_4/1 (4U)	10144	0.175	89.6	2.95
Q15C5_8/1 (6U)	9990	0.166	203.5	4.62

3.4. Piece 3 gas bubbler C-14 content

Each Piece 3 sample has four allocated bubbler activities as well as the C-14 content of the residual graphite. The oxidation times have been set in order to capture principally C-14 from the deposits in the first stage and principally C-14 from the graphite in the second stage. However the allocation of C-14 between deposit and graphite may require some correction. Two gas bubblers are nominally allocated to the C-14 of the deposit (but will include some activity from oxidised graphite) and from now on will be noted D1 and D2. Two further bubblers are nominally allocated to the underlying graphite (but may include C-14 from any remaining deposit) and will now be known as G1 and G2. Activity has been quoted in Bq (Table 6) and is the activity based on the total volume of the sample (100 ml per bubbler). C-14 was positively identified in 14 of the 20 gas bubbler samples analysed. In order to display the results in terms of C-14 activity in the deposit compared to underlying graphite the activities from D1 and D2 have been combined as have G1 and G2 (Figure 5). Less than values are quoted when a result falls below the level that can be determined as an exact activity. The Minimum Detectable Activity (MDA) is a function of the background (or blank) counts and is based on a multiple (4.65) of the standard deviation of the counts (i.e. $4.65\sigma_b$). The MDA formulas are based on the Currie paper [6] and documented in an American National Standard [7]. The less than values have been removed from any comparisons made and are only present in the second bubbler in the train, which is not unusual.

Similar to the Piece 1 C-14 activities, the J17A4 sample has the lowest activity compared to the other samples. Again the Q15C5 samples C-14 activity decreases as axial height in the core increases. The gas bubbler data shows that there is a higher activity of C-14 released from the deposits than there is from ~1-2% of the underlying graphite. However these results do not take into account the activity present in the remaining solid graphite samples.

TABLE 4 - PIECE 3 GAS BUBBLER ACTIVITIES

NNL Identity	C-14 Activity D1 (Bq)	C-14 Activity D2 (Bq)	C-14 Activity G1 (Bq)	C-14 Activity G2 (Bq)
J17A4_13/1 (9U)	5.51E+03	2.14E+00	1.63E+03	2.57E+00
Q15C5_2/1 (3U)	4.23E+04	<1.4E+00	8.88E+02	< 1.4E+00
Q15C5_3/1 (4L)	2.80E+04	<1.4E+00	6.40E+02	< 3.8E+00
Q15C5_4/1 (4U)	1.35E+04	2.27E+00	7.36E+02	< 3.7E+00
Q15C5_8/1 (6U)	1.01E+04	3.66E+00	1.44E+03	< 3.7E+00

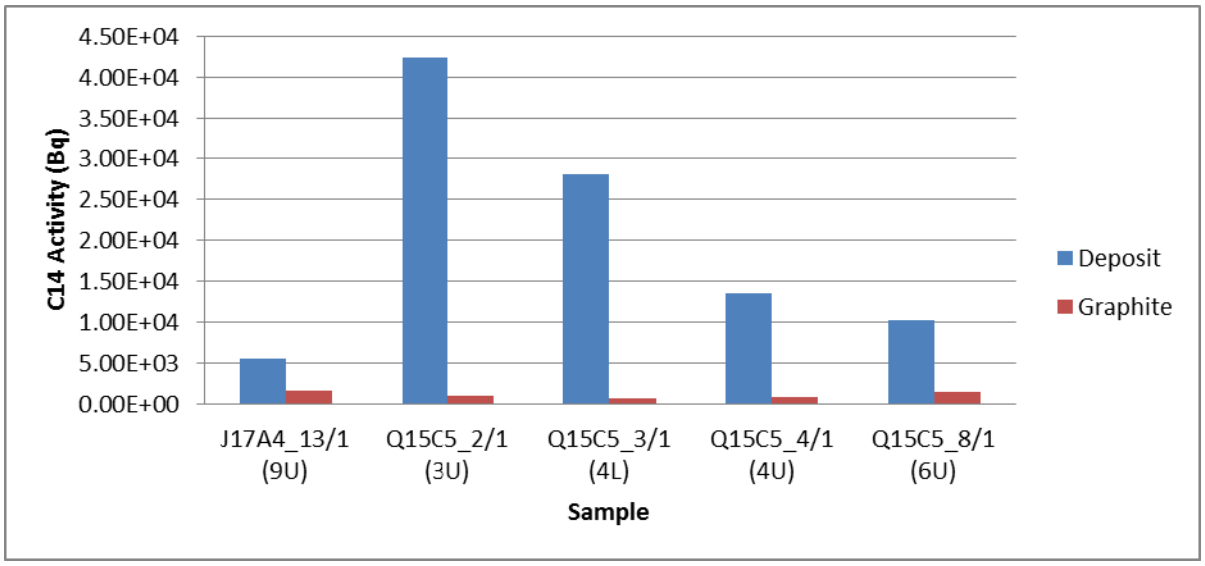

FIG.5. Piece 3 Gas Bubbler Activity

TABLE 5 - PIECE 3 C-14 CONTENT OF RESIDUAL GRAPHITE

NNL Identity	Weight (g)	C-14 activity (Bq/g)	Axial height in core (m)
J17A4_13/1 (9U)	0.190	8.03E+04	6.85
Q15C5_2/1 (3U)	0.211	6.03E+04	1.98
Q15C5_3/1 (4L)	0.205	5.57E+04	2.66
Q15C5_4/1 (4U)	0.363	6.71E+04	2.95

Q15C5_8/1 (6U)	0.368	8.83E+04	4.62

3.5. Piece 3 C-14 content of residual graphite

C-14 was positively identified in all of the Piece 3 residual graphite samples. Activity has been quoted in Bq/g (Table 7 and Figure 6). There is again a significant amount of variation between the samples. The activities ranged from 5.57E+04 Bq to 8.83E+04 Bq. Deviating from the trend is the J17A4 sample. While taken from the highest position in the core, this sample does not exhibit the lowest activity. But this sample comes from a different fuel channel where depositing conditions may differ.

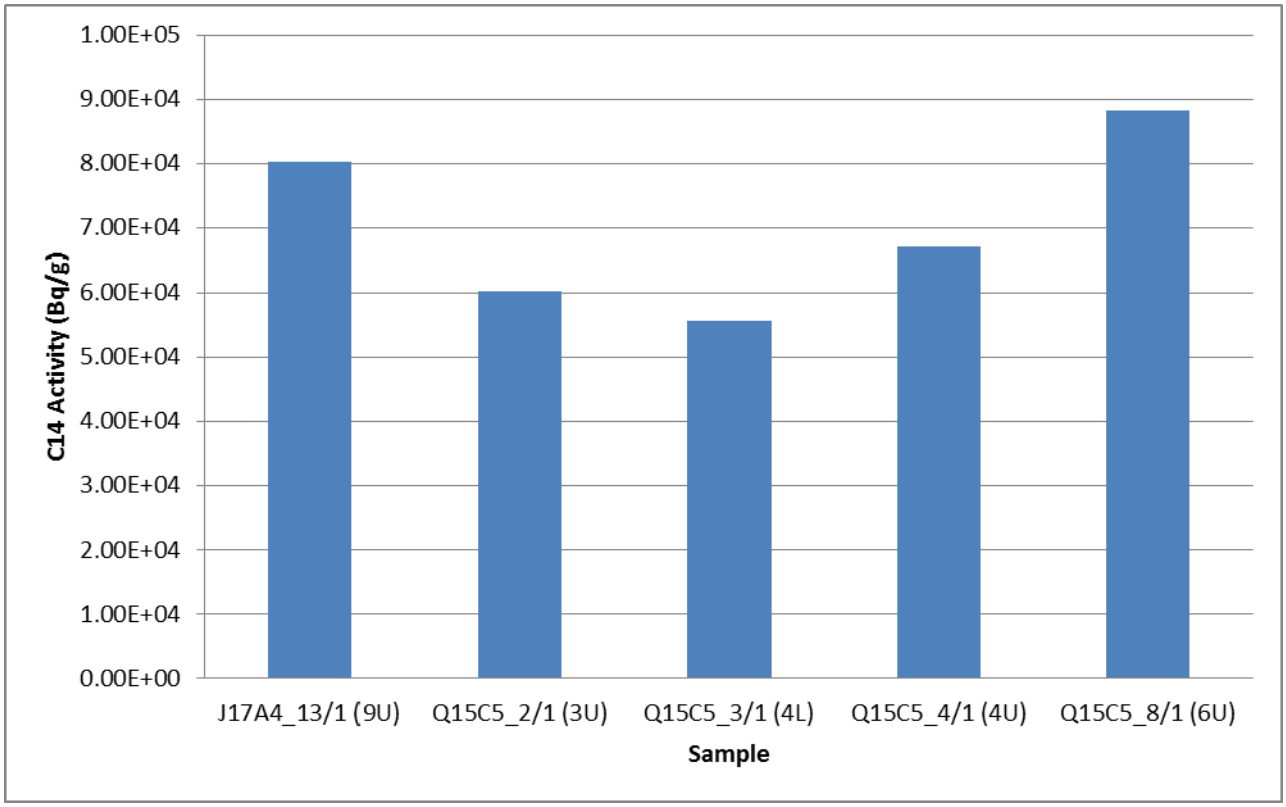

FIG.6. C-14 activity for Piece 3 residual graphite

3.6. Mass balance

A mass balance calculation was performed for each sample in order to compare activities from Piece 1 solid samples with the combined activities of the Piece 3 bubbler and graphite solid samples (Figure 7). Unfortunately, the mass balances ranged from 77-33%. This may be due to the fact that the level of deposit is not uniform throughout the whole disc sample or was changed during machining into three pieces. It may be coincidental that the discrepancy increases with increasing total C-14 activity.

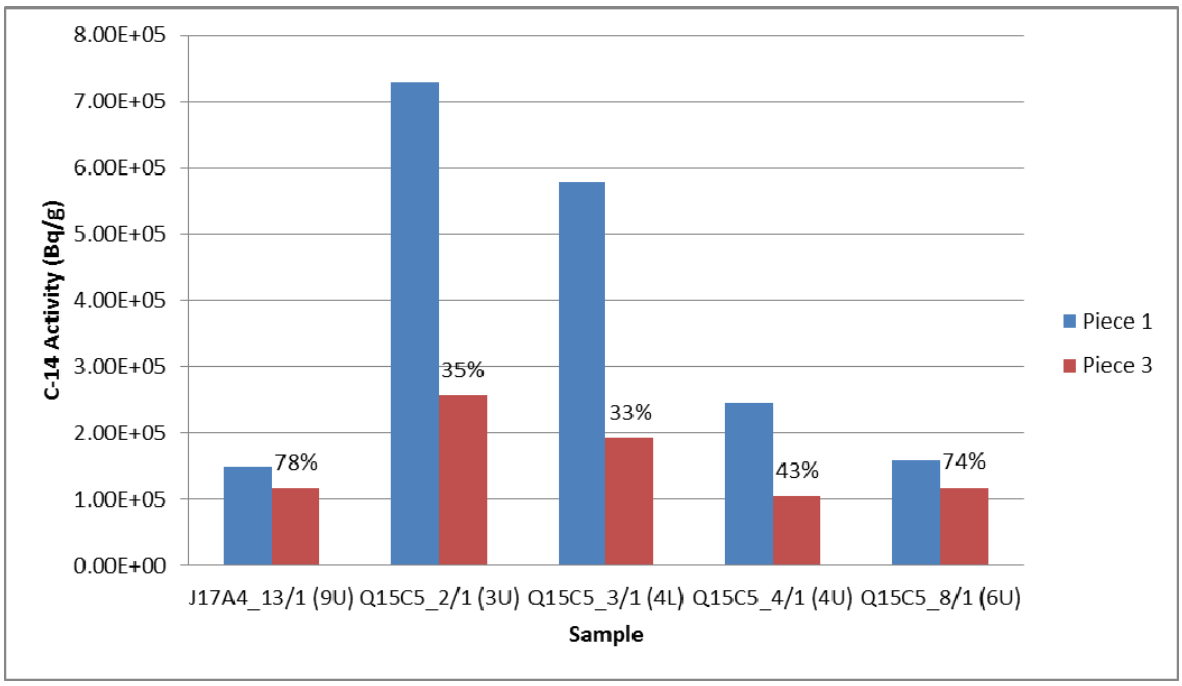

FIG.7. Mass balance

3.7. Extended oxidation

This experiment was included to measure the rate of C-14 released with increasing mass loss. After the deposit was removed, oxidation was continued until ~4% weight loss with the Dreschel bottles being changed at four ~0.5% weight loss increments. Sample J17C5_8/1 (6U) in Table 2 was chosen for this analysis. There were 10 Carbosorb gas bubblers associated with this experiment, two for each oxidation step. For illustrative purposes the data from both bubblers in one step have been combined (Table 8). As can be seen from Figure 8, there is significantly higher C-14 content in the bubblers attributed to the deposit than in the remaining bubblers combined. As mass loss increases the C-14 activity continues to decrease.

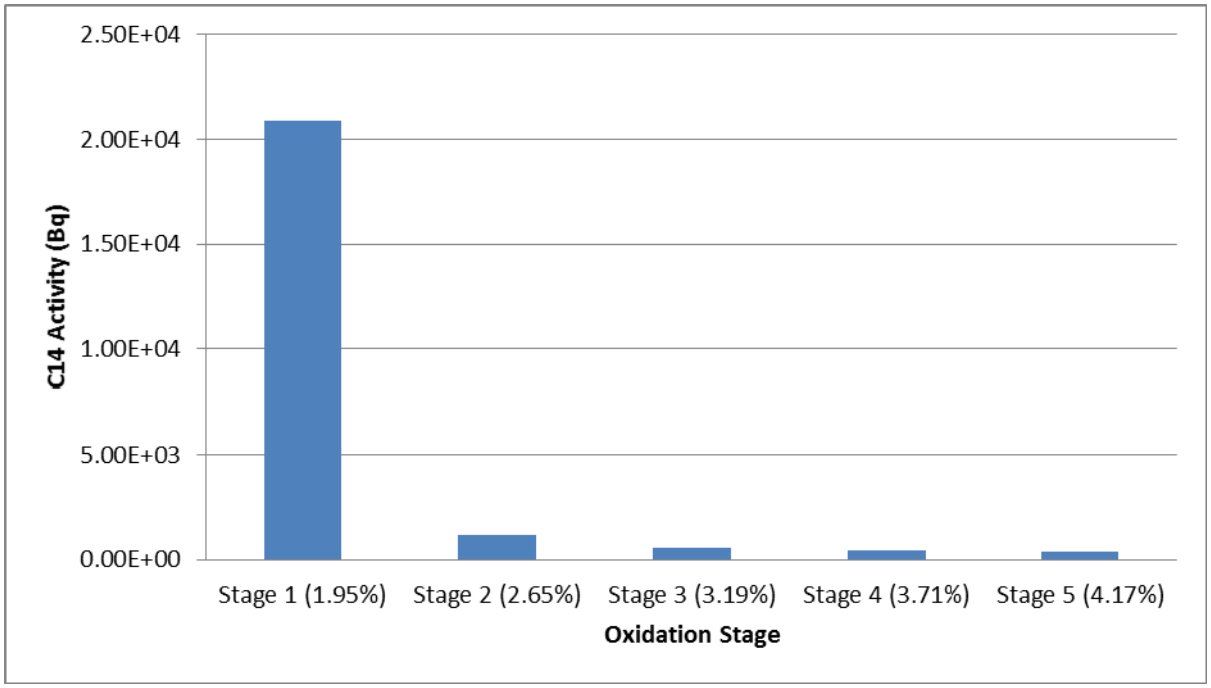

FIG.8. C-14 Activity with Increasing Mass Loss (values in brackets show cumulative weight loss)

TABLE 6 - C14 ACTIVITY WITH INCREASING MASS LOSS

Sample Name	C-14 Activity Bubbler 1 (Bq)	C-14 Activity Bubbler 2 (Bq)	Combined Activity (Bq)
Deposit	2.09E+04	1.63E+00	2.09E+04
Step 2	1.19E+03	1.47E+00	1.19E+03
Step 3	5.80E+02	2.50E+00	5.83E+02
Step 4	4.46E+02	1.80E+00	4.48E+02
Step 5	3.64E+02	2.12E+00	3.66E+02

3.8. C-14 correction

In order to accurately interpret the allocation of C-14 activity between deposit and graphite, it was necessary to perform a correction to the activity data from the gas bubblers. This is because the activity attributed to the deposit may contain activity from oxidised graphite and vice versa.

The specific activity of graphite used in the calculation was from the residual graphite remaining after TGA of Piece 3 samples. The specific activity of C-14 of the deposits had to be calculated by combining the TGA profiles of deposit and underlying graphite to extrapolate the mass of deposit and graphite lost. By multiplying the mass of graphite lost by the specific activity of graphite, the amount of C-14 activity attributed to graphite was obtained. Subtraction of this value from the total C-14 activity detected in the gas bubbler (nominally attributed to the deposit) provides a more accurate C-14 activity attributed to the deposit (Bq). Dividing this value by the extrapolated mass of deposit calculated, provides the specific activity of C-14 from the deposit (Bq/g) (Table 9).

TABLE 7 – CORRECTED C-14 ACTIVITY DATA

NNL Identity	Deposit C-14 Activity (Bq/g)	Graphite C-14 activity (Bq/g)
J17A4_13/1 (9U)	1.92E+06	8.03E+04
J17C5_8/1 (6U)	7.42E+06	7.14E+04
Q15C5_2/1 (3U)	9.78E+06	6.03E+04
Q15C5_3/1 (4L)	9.29E+06	5.57E+04
Q15C5_4/1 (4U)	4.67E+06	6.71E+04
Q15C5_8/1 (6U)	3.05E+06	8.83E+04

Specific activity of the deposit follows the same trend as previous data showing an increase in activity as axial height in the core increases. The average specific C-14 activity of graphite is 7.05E+04 Bq/g. Comparing this figure with the average specific C-14 activity of deposit which is 6.02E+06 Bq/g reveals an increase by of a factor of 85. These trends are not seen in the specific activity of the graphite, which is broadly similar for all samples.

3.9. Significance of C-14 levels in deposits

The analysis presented above, showing significantly higher C-14 specific activity in deposits compared to that for the underlying graphite, must be put into perspective in terms of C-14 activity across a whole fuel brick. Deposits are found principally on external brick surfaces in the fuel channels and within the graphite pores within a few millimetres of these external brick surfaces. A very approximate estimate of the total C-14 activity in deposits and in the underlying graphite can be made.

The mass of a fuel brick (average of an octagonal and square brick) is 89 kg taking no account of mass loss by radiolytic oxidation. Using the brick from which specimen Q15C5_2/1 (3U) was taken as an example, the total C-14 activity of the graphite will be 5.37E09 Bq based upon the residual activity in Table 7. Assuming a uniform deposit cover, the deposit mass for the whole brick can be estimated from the ratio of the areas of the entire fuel channel surface to that of specimen Q15C5_2/1 (3U) and the mass of deposit on the specimen calculated in the analysis of the oxidation data in section 3.8. The total mass of deposit is estimated to be ~50 g and so the total C-14 activity on the brick using the specific activity from Table 9 is 4.38E08 Bq. Therefore the C-14 from deposit accounts for ~8% of the total C-14 activity in the brick. It is possible that this is an under-estimate: bricks with interstitial channel cut-outs are known to have deposits on interstitial channel surfaces (at a lower concentration). There may also be deposits on other outer brick geometric surfaces where there is no face to face contact between bricks, but there are no data for this. The brick selected here for analysis lies ~2 m from the bottom of the core. It can be seen from Figure 9 that potential C-14 contribution from carbonaceous deposit will vary with axial position (the best-estimate dashed curve).

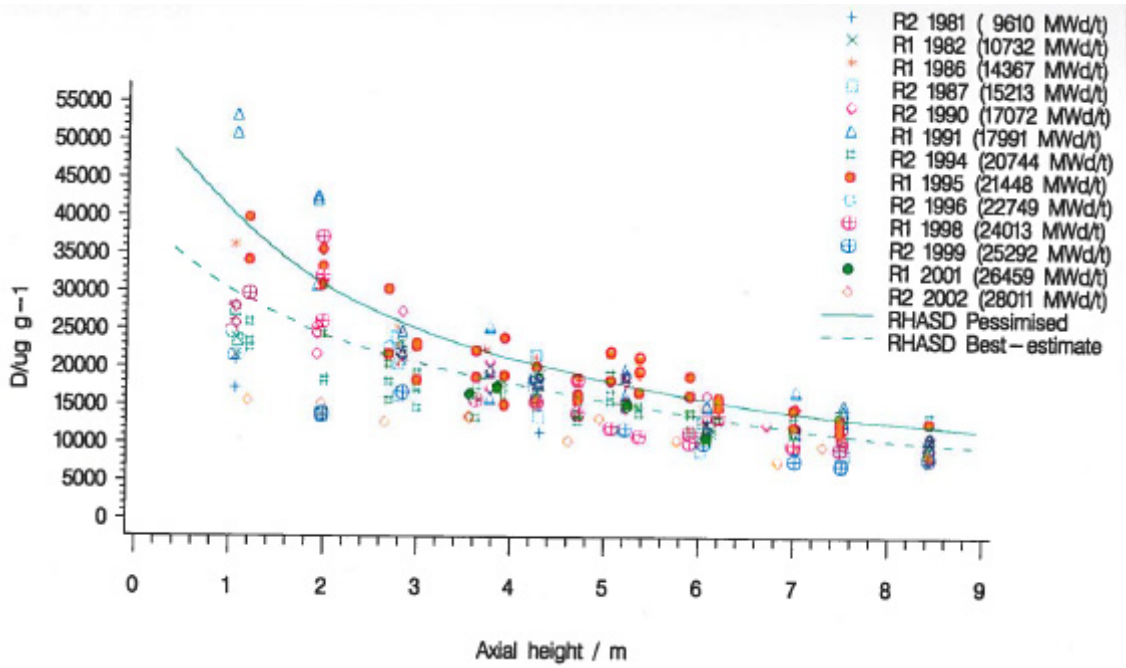

FIG.9. Axial Variation in Deposit Concentration at the Slice 1 Position in an Oldbury Fuel Channel

While some form of oxidation treatment to remove deposits would reduce the C-14 activity of core components, any benefits for final disposal of the graphite cannot be quantified from the limited analysis presented here.

4. CONCLUSIONS

The amount of deposit present on reactor graphite samples varies and appears to correspond with axial height in the reactor core, as previously observed in all Magnox cores. These deposits also have variable oxidation rates which may indicate the presence of catalytic impurities.

A significantly higher C-14 specific activity in the deposit was observed compared with that of the graphite. From the very limited data obtained from this study, the deposit specific activity showed a decrease with increasing axial height in the core whereas the graphite specific activity was broadly constant.

The variable (and in some cases poor) mass balances indicate that the level of deposit may not be uniform throughout the whole disc sample which were cut into three pieces.

The data from the extended oxidation experiment shows that the C-14 activity decreases significantly with increasing mass loss. The trend in C-14 activity after this first oxidation stage indicates the presence of some residual deposit in the second oxidation stage.

The average of the C-14 specific activities calculated for deposit and graphite indicate a significant difference, with the average C-14 activity of deposit higher than the average C-14 activity of graphite by a factor of 85.

Using a specimen taken from brick 3 as an example, the contribution from the deposit to the total C-14 activity of the whole fuel brick has been estimated to be ~8%. This contribution will vary with axial position within the core. While some form of oxidation treatment to remove deposits would reduce the C-14 activity of core components, any benefits for final disposal of the graphite cannot be quantified from the limited analysis presented here.

This study has been based upon just six graphite specimens taken from one Magnox reactor. Further work would be necessary to confirm the findings and to assess their significance for other cores in the Magnox fleet. Until such work has been undertaken, extreme caution should be taken in any application of the results from this study.

REFERENCES

[1] INTERNAIONAL ATOMIC ENERGY AGENCY, 'Progress in Radioactive Graphite Waste Management', *in:* Report of IAEA Technical Committee Meeting on Progress In Radioactive Graphite Waste Management, IAEA-TECDOC-1647, IAEA Vienna (2010)

[2] NUCLEAR DECOMMISSIONING AUTHORITY, 'The 2010 UK Radioactive Waste Inventory: Main Report (2010) (www.nda.gov.uk/documents).

[3] MARSDEN, B.J., HOPKINSON K.L., and WICKHAM A.J., 'The Chemical Form of Carbon-14 within Graphite'; Serco Assurance report SA/RJCB/RD03612001/R01 Issue 4 (March 2002) (www.nda.gov.uk/documents).

[4] MUGHABGHAB, S. F., 'Atlas of Neutron Resonances'; 5th Edition, Elsevier, (2006) ISBN: 978-0-444-52035-7.

[5] LAKER, A., 'UK Management of Solid Low Level Radioactive Waste from the Nuclear Industry: LLW Management Plan', Nuclear Decommissioning Agency (2009) (www.nda.gov.uk/documents).

[6] CURRIE, L.A., 'Limits for Qualitative Detection and Quantitative Determination, Application to Radiochemistry'; Analytical Chemistry, **40** (3) *pp.* 586-693 (1968)

[7] HEALTH PHYSICS SOCIETY, 'Performance Criteria for Radiobioassay', American National Standard, HPS N13.30-1996