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Radiocarbon mass balance for a Magnox nuclear power station

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ABSTRACT

Nuclear power generation in the United Kingdom is based principally on graphite-moderated gas-cooled reactors. The mass of irradiated graphite associated with these reactors, including material from associated experimental, prototype and plutonium production reactors, exceeds 96,000 tonnes. One of the principal long-lived radionuclides produced during graphite irradiation is radiocarbon (C-14). Its potential as a hazard must be taken into account in decommissioning and graphite waste management strategies. While C-14 production processes are well-understood, radionuclide distributions and concentrations need to be characterised. A common misconception is that generic statements can be made about C-14 precursors and their location. In fact, the composition of the original manufactured material, the chemical environment of the graphite during service and its irradiation history will all influence C-14 levels. The analysis presented here provides the first assessment of the principal C-14 activation pathways for a UK Magnox reactor. Activation modelling has been used to predict C-14 production rates in both the graphite core and the carbon dioxide coolant over a selected period of operation and the results compared with monitored site C-14 discharges. Principal activation routes have been identified, which should inform future graphite waste management strategies relating to radiocarbon.

1. Introduction

Nuclear power generation in the United Kingdom is based principally on graphite-moderated gas-cooled reactors. The mass of irradiated graphite associated with these reactors, including material from associated experimental, prototype and plutonium production reactors, exceeds 96,000 tonnes (The 2010 UK Radioactive Waste Inventory, 2011). One of the principal long-lived radionuclides produced during graphite irradiation is radiocarbon (C-14), which has a half-life of 5730 ± 40 years (JEF-2.2). Decommissioning and graphite waste management strategies must take account of this potentially hazardous radionuclide. In order to identify appropriate management options, it is important that the production processes and operational releases are understood, and the distributions and concentrations of C-14 are characterised.

In fact, C-14 precursors and their activation processes are well-known (see for example Marsden et al., 2002; IAEA-TECDOC-1521, 2006). However, there is ongoing debate over the relative importance of different C-14 precursors, which will determine the location of C-14 within graphite components and hence its mobility/response to treatment. A generally held misconception concerning C-14 in irradiated graphite is that generic statements can be made about its precursors and their location. C-14 location and activities will depend upon the composition of the original manufactured graphite (raw materials, impurities), graphite mass loss due to radiolytic oxidation, the chemical environment of the graphite during service and the irradiation history of the graphite. So, while there may be some similarities across, for example, carbon dioxide (CO_2) cooled graphite moderated designs (Magnox, Advanced Gas-Cooled (AGR), Uranium Naturel Graphite Gaz (UNGG)), any informed assessment of the C-14 inventory of a core would require more-precise characterisation.

The analysis presented here focuses on a UK Magnox reactor core, Reactor 1 at Wylfa Nuclear Power Station. The objective of the analysis is to present a full C-14 mass balance over a selected period of operation for which there are accurate C-14 site discharge records. The analysis presented here is the first assessment of this type that has been undertaken for a UK Magnox reactor. Activation modelling is used to predict C-14 production rates in both the graphite core and the CO_2 coolant. Predictions for the graphite are benchmarked against C-14 analyses undertaken on samples taken from the Wylfa cores. Given the uncertainties in the analysis, a precise mass balance between C-14 production and C-14 discharged to the atmosphere may not be possible. However, the analysis should be capable of identifying principal activation pathways, which should inform future graphite waste management strategies relating to radiocarbon.

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1.1. Formation of C-14

The radioactive isotope of carbon C-14 is produced in the atmosphere by the capture of thermal neutrons from cosmic rays by nitrogen atoms (specifically N-14). C-14 can also be produced through thermal neutron capture by the naturally occurring isotope of carbon (C-13) and by oxygen (O-17):

$$\text{N-14} + \text{n} \rightarrow \text{C-14} + \text{p}$$

$$\text{C-13} + \text{n} \rightarrow \text{C-14} + \gamma$$

$$\text{O-17} + \text{n} \rightarrow \text{C-14} + \alpha$$

In the case of radiocarbon produced in the atmosphere, this makes no contribution to C-14 levels in as-manufactured Magnox reactor graphite (PGA graphite) as this graphite is manufactured from fossil fuel products in which C-14 is fixed and will have decayed. C-14 in irradiated graphite will therefore come from N-14, C-13 and O-17 impurities in the material, noting the neutron capture cross-sections and isotopic abundances summarised in Table 1.

1.2. Monitoring C-14 discharges at licensed nuclear sites

The Environment Agency sets limits and levels on the discharges of gaseous and liquid radioactive waste from nuclear licensed sites in England and Wales (2012). These are based upon statutory guidance from the UK government, requiring operators to use "best available techniques" to minimise the generation and disposal of radioactive waste such that the resulting radiological impact to the public is brought down to levels that are as low as reasonably achievable. Limits are set for a rolling 12-month period, based on discharges determined on a monthly basis.

In the case of the Wylfa nuclear site, records of daily C-14 discharges (GBq) are maintained for each reactor. These are based upon C-14 activities (Bq m^{-3}) measured in the discharge route, which can be assigned to CO_2 releases from the pressure circuits via controlled "blowdowns" (to control reactor pressure) and from normal losses from the coolant circuits during operation.

For the purposes of this study, the period of operation between consecutive statutory outages on Reactor 1 commencing in September 2009 and ending in August 2011 has been selected. The C-14 discharge data together with the reactor thermal power for this period are summarised in Fig. 1. The data show a sharp rise in C-14 discharges as the reactor comes up to full power, after which discharges level off. The significant rise in the discharge in the final month corresponds to the depressurisation of the coolant circuits and the release of approximately 235 t of CO_2. It should be noted that following such an event, the reactor gas circuits are put through a series of CO_2 pressure/depressurisation cycles to purge the system of air prior to power generation. It is the data from Fig. 1 that will be used in the C-14 mass balance analysis. Activation modelling will be applied both to the graphite and to the carbon dioxide coolant gas to quantify contributions to the total C-14 inventory from each of these activation pathways.

Table 1
Thermal neutron "2200 m s^{-1}" capture cross-sections producing C-14 and the isotopic abundances of the precursors (Mughabghab, 2006).

Species	Capture cross-section leading to C-14 (Barns)	Isotopic abundance (%)
N-14	1.86 ± 0.03	99.63 (N-14:Nitrogen)
C-13	0.00137 ± 0.00004	1.11 (C-13:Carbon)
O-17	0.235 ± 0.010	0.037 (O-17:Oxygen)

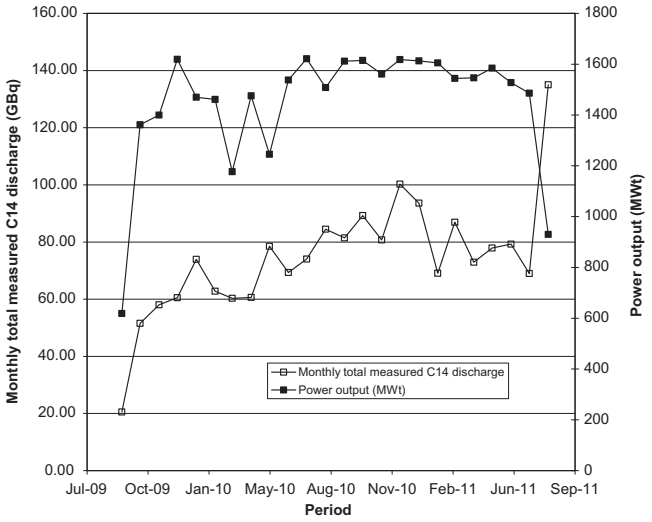

Fig. 1. Summary of C-14 monthly discharge data and associated reactor thermal power – Wylfa Reactor 1.

1.3. Previous related studies

An attempt to rationalise C-14 production in Magnox reactor cores with measurements in gaseous effluents has been reported by Doyle and Hammond (1980), where it was concluded that accurate C-14 predictions were impossible because of large uncertainties in impurity levels. The authors reported measurements taken at four Magnox sites, together with predictions specifically for Wylfa. Analysis of gaseous effluents provided C-14 activities (in MBq t^{-1} CO_2). Predicted C-14 concentrations in the coolant gas were reported, but the precise method employed was not described. The authors describe moderator source terms as C-13 and nitrogen in the moderator and CO_2 coolant source terms as C-13, oxygen and nitrogen impurity (50 volume parts per million). They noted that predictions were critically dependent upon nitrogen impurity levels and that a level of 7 weight parts per million (wppm) in the graphite would give a C-14 concentration in the coolant equivalent to that from C-13. The precise method for estimating this value is not explained, but appears to be a simple equating of reaction rates based upon estimates of thermal cross-sections, natural abundance and atomic weights. Using a similar method with current data and uncertainties from Table 1 above, a value of 8.8 ± 0.3 wppm can be calculated, assuming other C-14 producing nuclides (e.g. O-17) are not present. Such estimations do not take account of the full energy spectra of neutrons in the graphite of a Magnox reactor that vary both with location due to neutron transport and time due to graphite loss by oxidation and fuel burn-up effects (discussed further below). Effluent analysis for Wylfa Reactor 1 gave a measured C-14 activity in the coolant of 78 ± 4 MBq t^{-1} compared with a prediction of 44 MBq t^{-1}, with the difference being attributed to uncertainties in impurity levels.

Bush et al. (1984) reported a C-14 waste management study covering a large range of reactor systems including Magnox. Fluxes in various regions of an illustrative Magnox reactor core were used in conjunction with 2200 m s^{-1} cross sections and Westcott's method for adjusting cross-sections with temperature to perform activation calculations. Core parameters and impurity levels were selected on the basis of contemporary UK experience. The study concluded that C-14 arisings were dominated by the graphite moderator and specifically those from C-13 and N-14. The nitrogen impurity level was taken to be 10 wppm. At this level, their calculations showed that N-14 activation accounted for 61% of the total