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GRAPHITE MATERIALS TESTING IN THE ATR FOR LIFETIME MANAGEMENT OF MAGNOX REACTORS

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ABSTRACT

A major feature of the Magnox gas cooled reactor design is the graphite core, which acts as the moderator but also provides the physical structure for fuel, control rods, instrumentation and coolant gas channels. The lifetime of a graphite core is dependent upon two principal aging processes: irradiation damage and radiolytic oxidation. Irradiation damage from fast neutrons creates lattice defects leading to changes in physical and mechanical properties and the accumulation of stresses. Radiolytic oxidation is caused by the reaction of oxidizing species from the carbon dioxide coolant gas with the graphite, these species being produced by gamma radiation. Radiolytic oxidation reduces the density and hence the moderating capability of the graphite, but also reduces strength affecting the integrity of core components. In order to manage continued operation over the planned lifetimes of their power stations, BNFL needed to extend their database of the effects of these two phenomena on their graphite cores through an irradiation experiment. This paper will discuss the background, purpose, and the processes taken and planned (i.e. post irradiation examination) to ensure meaningful data on the graphite core material is obtained from the irradiation experiment.

1. Introduction

BNFL Magnox Generation Business Group in the United Kingdom is currently operating six commercial nuclear power stations, and is undertaking a program of work to underwrite continued operation of these nuclear power stations beyond their original design lifetimes. These are gas-cooled graphite-moderated reactors of the first generation Magnox design that utilize uranium metal fuel in magnesium alloy fuel cans. The reactor design has evolved as each power station in the series has been commissioned, with progressive increases in operating gas pressures and temperatures leading to increased power densities and thermal output.

For economic planning purposes, the reactors were designed with nominal lifetimes of 20-25 years. All the power stations are now operating well beyond their design lifetimes (30-40 years). In order to satisfy the operators and the United Kingdom regulators (HM Nuclear Installations Inspectorate) that the reactors continue to operate safely under normal operating and fault conditions, the reactors are subjected to rigorous periodic assessment. The graphite core of a Magnox reactor not only acts as the moderator which sustains the energy-releasing chain reaction but also serves as the main structural element of the core design, providing a lattice of vertical channels for fuel elements, instrumentation and flow of heat transfer gas. The assessment of the condition and behavior of the graphite moderator is a key part of this periodic safety review process.

With the graphite cores being operated significantly beyond their nominal lifetimes, it has been recognized that additional materials property data on irradiation damage and radiolytic oxidation are required to underwrite safety case assessments which will bound the projected irradiation and environment conditions. To this end, the Industry Management Committee within the United Kingdom (comprising British Energy, British Nuclear Fuels plc and HM Nuclear Installations Inspectorate) has provided funding for a graphite irradiation program. This program will irradiate samples of archive pre-characterized graphite used in the Magnox type reactors in the Advanced Test Reactor (ATR) at the United States Department of Energy's Idaho National Engineering and Environmental Laboratory (INEEL). After irradiation, the graphite will undergo Post Irradiation Examination (PIE) at BNFL Research and Technology's Berkeley Centre in the United Kingdom.

The new Irradiation Test Vehicle (ITV) facility in the ATR was selected for the irradiation to provide the desired irradiation conditions and on-line temperature control of the specimens. The ITV has the capability of irradiating specimens at elevated temperatures in a dry gas environment similar to an HTR. Depending on the amount and type of specimen and capsule material available for gamma heating, specimens may be irradiated in the ITV at temperatures as low as 200 °C or in excess of 800 °C. A new gas environment system was also added to the ITV to provide both oxidizing and inert gas atmospheres for the graphite specimens during irradiation. This will provide specimens with both the oxidation and neutron damage as well as specimens with just neutron damage effects. During PIE, the effect of each type of damage on the graphite can be then be assessed.

This paper discusses the irradiation experiment background, purpose and the processes taken and planned to ensure meaningful data on the graphite core material is obtained from the irradiation experiment.

2. Magnox Reactor Graphite Core Design and Operating Conditions

The cores are constructed from graphite bricks stacked in columns, geometrical stability being achieved through axial and lateral keying within the core in combination with a steel core restraint system. The columns of bricks are pierced by cooled vertical channels containing fuel elements, control rods and reactor instrumentation. The heat transfer medium is pressurized carbon dioxide. Pile Grade A (PGA) graphite, an orthotropic extruded needle-coke graphite, was chosen for the active regions of all the Magnox reactor cores, with a less pure graphite from an earlier stage of the same manufacturing process (Pile Grade B) being used for side reflector components.

Operating gas pressures are in the range ~8-27 bar absolute with operating gas temperatures of ~140-370°C. Reactor thermal powers range from ~270-1700MWt. The peak fast neutron dose to the graphite over the planned extended lifetimes of the stations is calculated to be in the region of 7×10^{21} neutrons/cm^2 Equivalent DIDO Nickel Dose (EDND).

3. Graphite Core Aging Process

There are two principal graphite aging processes: irradiation damage from fast neutrons creates lattice defects leading to dimensional changes and changes in properties including coefficient of thermal expansion, thermal conductivity, modulus and strength; exposure of graphite to carbon dioxide in the presence of gamma irradiation results in radiolytic graphite oxidation. This oxidation process produces changes to the porosity of the material leading principally to changes in modulus, strength and thermal conductivity.

The irradiation behavior of PGA graphite was extensively researched prior to and during the early development/construction period of the Magnox nuclear power stations. Fast neutron effects were studied using Materials Testing Reactors (MTR) principally in the United Kingdom (e.g. DIDO and PLUTO at Harwell, DFR at Dounreay) but also in France and Belgium (Siloe in Grenoble and BR-2 at

Mol). The temperatures and doses achieved in these studies bound Magnox station operating envelopes, even for extended operation. Irradiation damage in Magnox reactor graphite cores leads to a build-up of stored (Wigner) energy, distortion and dimensional change of components and, in combination with temperature and flux gradients, the generation of internal stresses within components. These processes are well understood and the MTR database for PGA graphite provides the means for predicting the effects of irradiation damage on core integrity.

Radiolytic graphite oxidation was studied in low dose experiments in MTRs in the United Kingdom (with samples being annealed to remove the effects of irradiation damage). The process involves the radiolysis of carbon dioxide to produce an oxidizing species. If these reactive oxidizing species impinge on a graphite surface, they gasify it to carbon monoxide. Depending upon the rate of diffusion of the oxidizing species within the pores of the graphite, it may become deactivated before it can react with carbon atoms in the graphite. The radiolytic oxidation of the graphite depends on the energy absorbed by the carbon dioxide (principally gamma) within the pores of the graphite. Oxidation rates increase with gas density – i.e. with increasing pressure and decreasing temperature. The presence of "inhibitors" such as carbon monoxide (a product of the process itself) and hydrogenous material (i.e. hydrogen, water or methane) will reduce oxidation rates. Data for the effects of radiolytic weight loss on PGA graphite properties are available up to weight losses of approximately 35%, with the database being somewhat sparse in the range 20-35% (Figure 1).

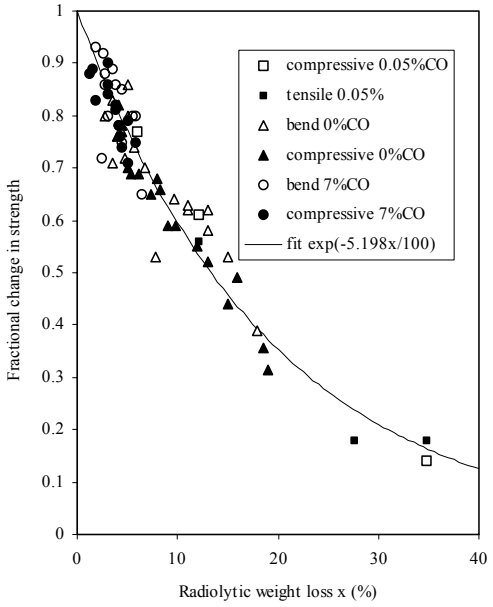

Figure 1: Fractional change in strength of PGA graphite with radiolytic weight loss

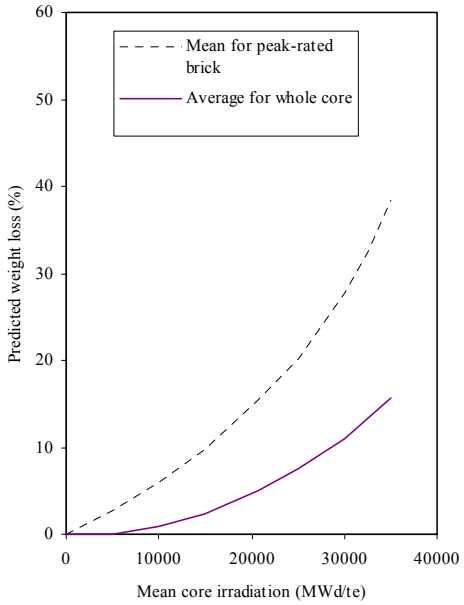

Figure 2: Radiolytic weight loss predictions for the Oldbury reactor graphite core

The adequacy of the database for radiolytic graphite oxidation must be assessed against measured and predicted weight losses in Magnox reactor core components. Figure 2 shows predicted weight losses for the Oldbury reactor core as a function of mean core irradiation. Weight losses have been expressed in two ways: as the average weight loss over the whole active core and as the mean weight loss for a brick in the peak-rated ("worst") position. The average weight loss of the graphite falls well within the database. For the small number of bricks at the peak-rated position within the core, mean weight losses are expected to lie at the database limit of 35% at a mean core irradiation of approximately 33,500 MWd te^{-1} (U). It is likely that part of the Oldbury core, the lead reactor on radiolytic graphite weight loss, will be operating during its planned lifetime outside the materials database unless new data from accelerated irradiation experiments can be acquired.

4. Objectives of Graphite Irradiation in ATR

There are two objectives for the graphite irradiation in ATR. The first and principal objective is the extension of the PGA graphite mechanical properties database to higher weight losses to bound reactor material over planned lifetimes for the Magnox nuclear power stations. Samples shall be irradiated in a controlled representative gas environment with target weight losses in the range 20-60%. The second objective is to crosscheck fast neutron damage effects between ATR and the existing MTR database for PGA graphite. In this case, samples shall be irradiated in an inert environment with instrumentation providing the accurate measurement of temperatures and doses.

5. ATR Facilities and Capabilities

The ATR's newly installed Irradiation Test Vehicle (ITV) was chosen as the position to perform the irradiation. The ATR is a premiere MTR capable of accommodating large irradiation specimens (11.5 cm in diameter by 1.22 m long) within very high neutron flux (1 x 10^{15} neutrons/cm^2/sec thermal and 5 x 10^{14} neutrons/cm^2/sec E $\geq$ 1MeV) and gamma heating (~ 20 W/g in stainless steel) environments. The ITV is located at the very center of the ATR core and is an extremely flexible facility designed to support a variety of experiments requiring fast, thermal, or mixed spectrum neutron environments. This facility is capable of providing neutron spectral tailoring and continuous temperature control for up to 15 capsules with inside diameters of 1.35 to 2.21 cm and lengths of 15.2 to 35.6 cm. Smaller numbers of longer length capsules can also be accommodated.

The in-reactor portion of the ATR ITV consists of three closely packed mini-in-pile tubes (MIPTs) running the length of the ATR vessel. The inside bores of the MIPTs are kept dry and test trains are inserted from the top of the reactor into the dry cavities. Temperature control gas lines enter through the bottom of the MIPTs and remain in place between tests. The temperature measurement and any other desired instrumentation or support systems are supplied with the test train and connections are made to the control system at the ATR vessel top head area each time a test train is replaced. Neutron spectral tailoring is accomplished through the use of replaceable filters that are inserted between the reactor fuel and an aluminum filler piece surrounding the MIPTs in the core region.

Independent temperature control of each capsule within the ITV is accomplished by controlling the transfer of the gamma and neutron heating of the capsule and its contents across a narrow gas jacket to the ATR primary coolant. During operation, the precisely sized gas jacket between each capsule and the mini-in-pile tube receives a continuous supply of gas custom blended for heat transfer purposes. By adjusting the ratio of a conductor (helium) gas and an insulator (neon) gas, the thermal conductivity of the gas jacket can be controlled to provide the correct insulating value between the nuclear heated capsules and the relatively cold (60 °C) reactor primary coolant. This controlled heat transfer drives the temperature of the specimens to the desired values during reactor operation. The correct temperature is maintained in each capsule by blending the mixture ratio of the two gases based upon temperature feedback from thermocouples located within each capsule. The blending operations are completely automated and are performed using Mass Flow Controllers (MFCs) that are linked to a computer control console in a manned experiment operation center. The exhaust lines leave the MIPTs at the reactor bottom head, pass through a leak detection panel, and discharge to the reactor ventilation system, which exhausts to the ATR stack.

6. Experiment Description

The graphite irradiation will be performed in MIPT #3, which is in the west position of the ITV. The experiment will consist of a single test train, which will be split into two capsules containing the Magnox graphite specimens. The two capsules are of equal length, separated at the vertical center of the ATR core by a sealed bulkhead, and extend from the center to the upper and lower ends of the active fuelled region of the reactor core. The top capsule, with a flowing inert (helium) gas environment, will contain fast neutron damage control specimens and the bottom capsule, with a

flowing oxidizing (CO_2) gas environment, will contain specimens to be radiolytically oxidized to high weight losses. In both capsules, the graphite will be positioned within a specimen carrier to ensure the applicable gas flow is positively directed over as much of the specimen surface as possible. This was done to minimize the diffusion distance for the gas within the porosity of the graphite specimens. To ensure data from the irradiation specimens would be directly comparable to data obtained from samples trepanned from Magnox reactor cores, the same standard specimen size was used in the irradiation. Other prototypic conditions mandated for the irradiation included specimen temperatures and gas environment (gas composition and gas pressure) of Magnox reactors. The length of irradiation was determined using a radiolytic oxidation model developed in support of the Magnox reactors. This model takes into account the total nuclear (gamma plus neutron) heating of the specimens, the composition of the gas and the desired weight loss of the specimens. Using this input, the model predicted approximately 140 irradiation days in the ATR ITV would lead to radiolytic weight losses which would bound those calculated to arise over the planned lifetimes (including planned life extension) of the Magnox power reactors. If the oxidation specimens reach the desired amount of oxidation prior to the end of the irradiation cycle, the oxidation system will be flushed with an inert (helium) gas and then purged with the same gas throughout the remainder of the irradiation cycle.

Extreme care was taken in the design of the experimental hardware to maintain prototypic chemistry control of the specimen environment to limit the unknown variables and prevent any unwanted chemical reactions during the irradiation. Materials that could either affect (e.g. catalyze or block) or mask the oxidation rate (e.g. carbon deposition) were excluded from the experimental hardware and all communicating systems. The major component of most ATR test trains and capsule shells, stainless steel, was excluded due to its nickel content. To satisfy the chemistry requirements, the capsule material was researched extensively and 9Cr 1Mo alloy steel was selected. This material was able to meet the chemistry requirements and also survive the high neutron flux environment of the ATR without experiencing a significant increase in its brittle fracture transition temperature. However, since the design code required a post weld heat treatment for this material, special techniques were needed to prevent melting of the aluminum specimen carrier during this process.

A vertical section of the experimental hardware is shown in Figure 3, and a horizontal cross-section at the top of the capsules is shown in Figure 4. The specimen carrier consists of two clamshell halves that are held together by thin aluminum straps. These straps also hold the thermocouple leads, gas lines and flux monitors in grooves machined in the outer surface of the carrier. Aluminum was chosen for the specimen carrier material for several reasons including chemistry control, low neutron absorption rate, high thermal conductivity and a high thermal expansion rate. The last two properties enable the carrier to provide an excellent heat transfer path between the specimens and the capsule wall. The high thermal expansion rate also helps in the assembly and later disassembly of the capsules by providing needed clearance between the carrier and the capsule wall at room temperature. However, during irradiation the aluminum expands to provide positive thermal contact between the carrier and the

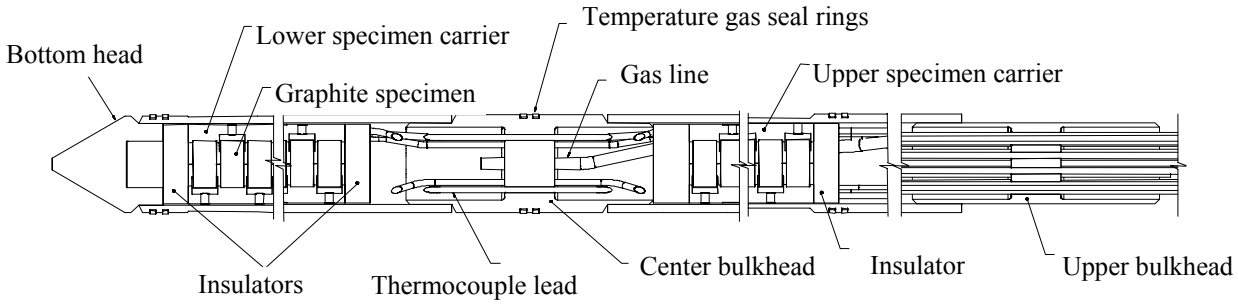

Figure 3: Vertical section of the experiment capsule showing the orientation of the capsules and the bulkhead separating the two capsules.

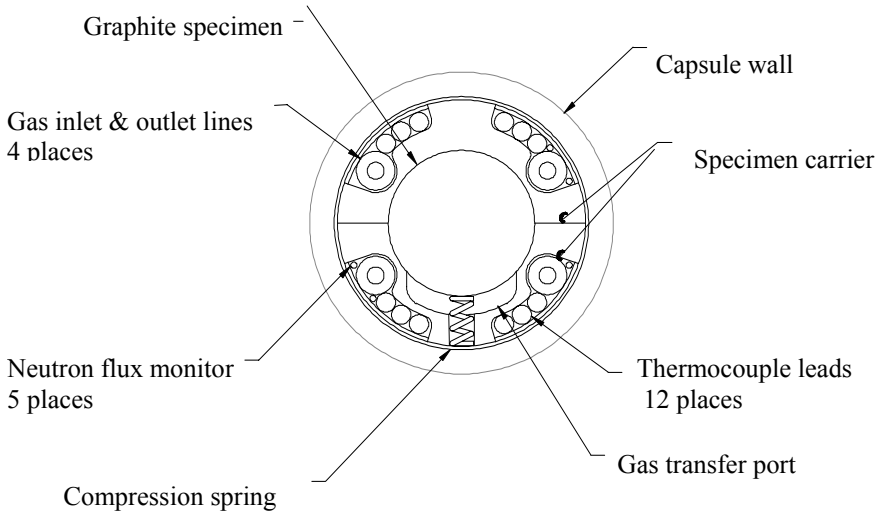

Figure 4: Cross-section of the experiment capsule showing the graphite specimens located in the center of the capsule and surrounded by the specimen carrier and other key components.

capsule wall. Nonetheless, the carrier's main role is to provide support for the graphite specimens, whilst separating them by a 1-mm gas space between adjacent top and bottom surfaces of the specimens. The gas spaces (or gas path) between the specimens are connected by gas ports located on alternating sides of the specimens. The specimens are held against the side of the carrier opposite the gas port by a compression spring to ensure a good thermal contact between the specimen and the carrier for heat transfer purposes. Thermocouples are located at the top, middle and bottom of each capsule for both temperature control and recording the temperature of the specimens. At each of these locations there are two thermocouple junctions located diametrically opposite each other: one measuring the centerline temperature of a graphite specimen and one measuring the temperature of the specimen carrier. Neutron flux monitors were installed to measure both the fast and thermal neutron fluences. Iron, nickel, and niobium wires measure the fast spectrum, whilst cobalt-aluminum alloy wires measure the thermal spectrum. Two iron flux monitors were also positioned diametrically opposite each other in each capsule to measure the fast neutron flux gradient across the capsules. Bare monitor wires were utilized in the upper inert capsule, but to prevent any unknown chemical effects on the specimens or wires, the monitors were encapsulated in a sealed aluminum tube in the lower oxidation capsule. Since each capsule has a different gas environment, there are separate gas inlet and outlet lines for each capsule. The inlet gas lines terminate at the bottom and the outlet gas lines are located at the top of the capsules. The gas (either oxidizing or inert) enters the gas plenum from the inlet gas line at the bottom of each capsule. It then traverses the tortuous path up between the specimens via the spaces between the specimens and the gas ports to enter the outlet line at the top of the capsule.

The outside diameter and profile of the capsule was determined based upon very rigorous reactor physics and thermal analyses. In order to ensure the data from this experiment was directly comparable with the existing database, an equivalent flux and temperature relationship established using data from the DIDO reactor was used to determine the required specimen temperatures. This relationship was applied to the neutron flux profile of the ATR to determine the correct temperature of the specimens based upon their axial position within the ATR core. The outside diameter of the capsule wall was then tapered to provide the correct insulating gas jacket between the capsule and the MIPT as a function of vertical location within the ATR core.

The capsule environment system is a new capability added to the ITV specifically for this project. It consists of the two different gas systems (inert and oxidizing) connected to the inert and oxidizing capsules in the MIPT. The inert gas system contains pure helium gas, and the oxidizing gas system contains a carbon dioxide gas that is blended with a carbon monoxide/hydrogen mixture. A

description of the requirements for and the design of the complex control system are beyond the scope of this paper. Inlet and outlet gas compositions are monitored over the duration of the experiment using a gas chromatograph and oxidizing gas flow rates and composition adjusted to take account of the oxidation of the graphite.

7. BNFL Facilities and Capabilities

BNFL currently operates hot cell facilities in the UK at Berkeley and at Sellafield. The main graphite facilities at Berkeley comprise a suite of cells and glove boxes for medium and low active work. Berkeley has been involved in the characterization of graphites in the UK for many years and has developed considerable expertise and designed specialized equipment for such investigations. Techniques and methods available for irradiated graphite include: precision machining equipment and equipment for mensuration, weighing, high temperature annealing, volume determination, helium pycnometry, strength, dynamic Young's modulus, coefficient of thermal expansion, diffusivity, permeability, thermal conductivity, stored energy measurements and chemical behavior. In addition, a wide range of optical and electron microscopes are available including a shielded scanning electron microscope. The Berkeley capability also includes surface and image analysis instruments.

8. Characterization of Material for Irradiation

Archive graphite retained during the construction of the reactor cores has been used as the source of material for the experiment. For the purposes of the experiment described here, cylinders of graphite were machined from selected heats of archive graphite with the grain direction either perpendicular or parallel to the axis. The cylinders were sectioned to provide discs 12.00mm in diameter and 6.00mm in length (machining tolerances of ∀0.03mm). Each disc either side of the disc destined for the experiment has been retained as reference material. Standard waisted tensile strength samples were also machined from selected sections of the cylinders for reference. All the graphite discs were subjected to detailed mensuration and weighing and measurement of dynamic Young's modulus. Selected graphite discs were subjected to open-pore volume (helium pycnometry) and coefficient of thermal expansion measurements. All the data were subjected to detailed analysis, with samples with outlying properties being examined for microcracks/flaws and rejected as appropriate. For each heat and for each grain orientation, samples were then paired to match properties. Samples from each pair have been assigned to the oxidation and inert irradiation capsules at symmetrical positions about the center of the ATR core to provide identical irradiation conditions.

9. Summary

Extreme care has been taken in the development of the irradiation experiment to ensure the data will be directly comparable to the existing database. The design of the experiment test train and the capsule environment system has taken all known variables into account and every effort was made to minimize the effects of any items (e.g. chrome, gold, etc.) that were unavoidable. This philosophy together with the nuclear heating and temperature control provided by the ATR ITV facility will provide the irradiation specimens needed to extend the current database on radiolytic oxidation of PGA graphite. This database may then be utilized in underwriting the safety cases for safe continued operation of the Magnox power stations.