Large and
ASSOCIATES
Consulting Engineers

MAGNOX DECOMMISSIONING DIALOGUE TIMESCALES WORKING TWG

REVIEW OF THE POSSIBILITY OF GRAPHITE CORE DEGRADATION DURING THE "CARE & MAINTENANCE" AND "SAFESTORE" DEFERRAL PERIODS AND DISPOSAL OPTIONS THEREAFTER

CLIENT: THE ENVIRONMENT COUNCIL

REPORT REF: R3069-A5

23 DECEMBER 2008

ABSTRACT

Most probably, the United Kingdom has the largest mass of irradiated graphite for storage and eventual disposal worldwide. This arises because of its early commitment to the graphite moderated reactor, first for breeding fissile plutonium for its nuclear weapons programme, then with the pioneering Magnox nuclear power station programme and subsequent generation of Advanced Gas Cooled Reactors. This paper reviews the knowledge and state of preparations for dealing with the deferred storage, dismantling, packaging and eventual disposal of the graphite moderators from the Magnox power stations.

Preparations for graphite related aspects of Magnox decommissioning, according to publicly available papers, seems to be confined to a single review of 1995, or thereabouts, that concludes in 1996 that little further R&D work is required to provide sufficient confidence in the proposed 10 year *Care & Maintenance* period followed by a 100 or more years of *Safestore*. The 1996 concluding report by Wickham and Marsden, et al, although authoritative, leans heavily on previous work all of which, apart from a few papers on the WAGR decommissioning programme, was directed towards the earlier R&D in developing (and maintaining) graphite for the role of moderator in the Magnox and later AGR operational reactors, and there is much extrapolation in applying these and the results gleaned from other reactor systems (and operating conditions) to the Magnox decommissioning programme.

This Review considers the main characteristics of graphite, as identified by Wickham and others, that are available for degradation during the extended *Care & Maintenance* and *Safestore* periods. It is agreed that providing the targeted passive *conditions* of *Safestore* can be achieved and maintained, then it is unlikely that graphite, in itself, will present any major problems for the safety of the reactor hulks over the time scales projected. However, the Wickham work does not consider in any great depth the extreme conditions that might arise at the introduction of an external (or internal) hazard, so the work is exclusive of a full hazard/fault analysis, which might be considered to be a shortcoming in the graphite chapter of the nuclear safety case for decommissioning.

Specifically, this Review identifies a number of areas where the longer term graphite performance (durability) should be tested against reasonably foreseeable hazards and that it may be prudent to assess the impact of graphite leaching on a station by station basis for each of the *Safestore* time scale options under consideration.

Large & Associates The Gatehouse 1-2 Repository Road Ha Ha Rd Woolwich Common London UK SE18 4BQ

☎ 44[0]20 8317 2860 *fax* 44[0]20 8317 2859 *mobile* 07971 088 086 mailto:largeassociates@msn.com

GRAPHITE DURABILITY DURING *CARE & MAINTENANCE* AND *SAFESTORE PERIODS*

The UK nuclear industry has, possibly, the largest number of graphite moderated reactors worldwide.[‡] Although, except for Sizewell B, all of these reactors have graphite cores, for deferred decommissioning, eventual dismantling and disposal of the graphite a clear distinction should be made between the low temperature R&D units and the reactors of the commercial nuclear power stations.

Here we are concerned only with the earlier group of commercial nuclear power stations, the Magnox reactors and the condition of the graphite core at the time of the final reactor shut down, how and the extent to which the graphite core might further degrade over the projected *Care & Maintenance* (*C&M*) and *Safestore* periods, and how the graphite might withstand the environmental conditions imposed by the *Safestore* design and all foreseeable events that could intrude into the *Safestore* regime.

For both *C&M* and *Safestore* regimes conditions within and about the reactor core will be, essentially, passive for the anticipated storage conditions. However, conditions and circumstances could depart from the projected '*passive* 'envelope should an external or internal hazard event occur.

The boundaries of the projected *passive* envelope might be defined to be those of ambient environmental conditions (air flow, temperature and humidity), a degree of water intrusion, and loads and forces being imposed upon the graphite core structure by its own selfweight and that the surrounding structure. Abnormal or '*adverse*' conditions might arise that could introduce seismic shock, localised high temperature, explosion, water deluge, structural collapse of the core and/or the surrounding structure, and/or the introduction flammable substances and ignition of the graphite.

EXTANT CONDITIONS OF THE CORES - MONITORING AND SOURCE DATA

Over the years of operation all of the Magnox reactor cores, their support and restraint systems have been monitored so that a comprehensive volume of information is available to the operator. This information relates to the physical condition of the graphite, the history of irradiation from reactor to reactor, and the radioactive inventories,[1,2] for those reactors already shut and those waiting shut down.

The general observation is that it is the corrosion products of the non-graphite components within the cores (the steel restraint garter, etc), rather than the graphite itself, that could potentially give rise to an immediate radioactive release should the containment be breached. So in this respect, there is an acknowledged potential for radioactive release during the *C&M*

[‡] The early research and development reactors at Harwell (Gleep, Bepo, Hero, Horace, Zenith), Dragon at Winfrith, and the Windscale plutonium production piles and linked to these the subsequent generation of commercial Magnox, the development Windscale Advanced Gas Cooled Reactor (WAGR) and its commercial AGR variants were all graphite moderated.

and *Safestore* periods should the steelwork structures weaken to a point of collapse or a sufficiently severe *adverse* event occur. It follows that if corrosion ekes more into the structural integrity of the core restraint and support structures as time passes, then the potential for a core collapse increases with an extended *Safestore* period.

Setting aside the release of radioactivity from the corroding steelwork structures, the model for radioactive release from the graphite must include for relatively leisurely release modes, such as leaching which is considered later, and very disruptive events which challenge the physical strength and durability of the graphite in situ.

Magnox Electric claim that the graphite cores and supporting structures, that is the generic design and condition (durability) of the graphite core, is satisfactory[3] for the *passive C&M* and *Safestore* periods. Howsever, anomalies exist at individual power stations. For example, localised failure of the core restraint system has been identified at Dungeness,[4] high levels of carbonaceous dust at Sizewell, core keyways failures predicted for Oldbury,[5] and there is a potential for tensile cracking at one of the Calder Hall reactors. These station by station anomalies could be of concern within the operational safety regime, particularly during rapid depressurisation and/or steam ingress incidents, but should not be a significant factor during the C*&M* and *Safestore* periods, if that is, the individual station *Safestore* design and maintenance strategy takes account of any potential development of any extant anomaly.[§]

So far as the time scales of decommissioning relate, it might be that the accelerated development of one such anomaly might bring a particular power station forward in the final dismantling plan. It should be possible to identify such a requirement during the *C&M* period for each station.

WIGNER ENERGY

The accumulation of Wigner energy has been extensively studied for low, moderate and high temperature graphite moderated reactors.[6] The rate and accumulation of Wigner energy during irradiation (reactor operation) is significant in low temperature reactors, such as the Windscale piles, and subsequent rapid release of this energy may create difficulties during decommissioning, packaging and disposal of the graphite. Once a reactor has ceased operation there is no further accumulation of stored energy and in the longer term a small amount of the stored energy may naturally dissipate out.

The energy storage rate for moderate and higher temperature reactors, such as the Magnox reactors, is considerably slowed, and the total accumulation of energy is much lower, so much so, that the release of residual Wigner energy during the *C&M* and *Safestore* periods should be of no consequence if, for some adverse reason, the elevated temperature conditions under which an energy release could occur arose.

[§] Although not considered here in any great detail, regard has to be given to the final means of removal of the graphite for the containment at the time of final dismantling and, of course the durability and longer term performance is important in determining this optima of the time scale. Other factors may intervene that relate to the containment structure itself, for example there are believed to be difficulties and holds up in progressing the core dismantling of the Windscale No 1 pile because the biological shield roof is now of insufficient strength to mount the cutting and lifting equipment necessary to remove the graphite core remotely.

As a further safeguard, following the analysis of Windscale accident of 1957, operation of the Magnox reactors has been maintained below the 80% specific heat capacity defined by Fleck.[7]

GRAPHITE OXIDATION IN AIR

It is generally accepted that oxidation[**] of high quality, reactor grade graphite in air is effectively precluded below a temperature of 350°C, which by far exceeds the projected *passive* (ambient) temperature envelope of *C&M* and *Safestore*.

However, the radiolytic oxidation threshold could be significantly lowered if, for some reason, the reactor core remained fully fuelled for long periods whilst immersed in air, although once defuelled, the residual radiation field is of no consequence in this respect even for periods extending over 100 years.[8] It is believed that the Magnox reactors are to be finally defuelled under CO_2 within 3 to 4 years of shut down thus obviating this concern.

The inclusion of catalysts from certain types of contamination, particularly inorganic impurities which may be introduced to the graphite during normal operation of the reactor and during the prolonged shut down of the *C&M* and *Safestore* periods, might also lower the activation energy of oxidation. Such catalysts could be introduced by further corrosion of the steelwork, chemical breakdown of the concrete biological shield structures, and from other pathways linking the natural environment (air and water - including lead, sea salts and calcium). A level of catalytic contamination would have been established from normal operation of the reactors over their respective lifetimes.

For *C&M* and *Safestore* the intention is to control the natural air ventilation rates, thereby limiting the route of delivery of air entrained sea salt, and it is assumed that some form of barrier will be maintained between water/moisture linkage between the graphite core and the principal source of calcium, the biological shield.

Oxidation of graphite in air is well understood and under the projected *passive* conditions for the *C&M* and *Safestore* periods the risk of high rates of heat liberating oxidation in air should be disregarded. However, there is potential for reducing the oxidation temperature by the introduction of pollutant sourced catalysts via atmospheric air and water pathways introduced in the (longer term) aftermath of an abnormal event or in instances where the barriers have broken down by unforeseen and/or undetected degradation of the containments. The potential to introduce a large quantity of an activation energy reducing catalyst via some external event (ie aircraft crash) should be exhaustively studied.

Previous studies[9] have suggested there to be a potential for combustion risk in the WAGR core dismantling phase if an extraneous source of heat was introduced and held (ie an incident involving a thermic lance or BROCO). There is also some uncertainty over the oxidation performance when the graphite had been subject to a damp CO_2 atmosphere or soaked in water, for which further work may be required before this doubt can be fully resolved.[10]

[**] Consider graphite *oxidation* to mean an exothermic reaction and ignition, ie burning. Oxidation commences at an activation energy. The activation energy for graphite oxidation is important because a quite small increase can significantly increase the reaction rate and, hence, heat input at high temperature.

CARBONACEOUS DUSTS

Carbonaceous deposits will oxidise much more easily than the graphite itself and there will be opportunity oxidation in advance of graphite oxidation. The quantity of carbonaceous deposits is, for most Magnox reactors, believed to be at an acceptable level, although the levels within Sizewell A reactors may require some additional consideration for the *C&M* and *Safestore* periods.

Other than relatively small scale friction generating and abrasive movements within the core, there is little potential during the *C&M* and *Safestore* periods to generate more dust.

NITRIC ACID

Irradiation of moist air can give rise to the formation of nitric acid which could attack the ordered structure of graphite, although nitric acid levels found in operational and recently shut down Magnox units indicates this not to be a problem. The residual level of radiation in a defuelled reactor is insufficient to give rise to significant quantities of nitric acid over the *C&M* and *Safestore* periods.

BREAKDOWN OF THE GRAPHITE RADIOACTIVITY

To release the radioactivity of the graphite itself[††] the nominated mechanisms are leaching via water intrusion; gassing from the graphite, including for elevated temperature and biological action; and by mechanical means such as movement abrasion or from more energetic events, for example core collapse.

Water in contact with the core graphite during *passive C&M* and *Safestore* conditions will most probably derive from intruding rainwater and condensate. The chemical reactivity of this water will be conditioned by the interactions undergone in its path to the graphite and with the graphite itself, and the final quality of the leachate water will determine the solubility, and hence the release rates, of the individual radionuclide.

Also, the impact of leaching of radionuclides from the graphite core on the surrounding environment may be very sensitive to the particular station (and indeed the particular reactor because of its operating history and any variations in the graphite from reactor to reactor) and the local geology/hydrology. In this respect it is not clear whether Magnox Electric have undertaken modelling of the leachate impact for the individual Magnox reactors over the range of *Safestore* periods being considered as options – it may not be entirely justified to assume that there will be no ingress of water into or release of radioactivity from the containment over the *Safestore* period.

At *Safestore* conditions (ie ambient), gas-phase release from the graphite is found to mainly involve the liberation of tritium (H-3) which, in account of the relatively short half-life, may not be a longer term concern, although in the shorter term such ventilated discharges might

---

[††] The principal graphite isotope is carbon-14, the entrained activation products include cobolt-60, and fission products, such as caesium-137, that may have transferred from the fuel during operation.

breach acceptable levels of atmospheric discharge through the ‘breathing’ arrangements to be put in place for *Safestore*.

Other than mechanical abrasion cause by small differential movements within the graphite core during the *Safestore* period, and excluding a catastrophic event to the containment and structure within, the potential for release of radionuclides bound into or attached to the graphite via this mode is limited. As previously discussed a core collapse within a sound containment would most likely not result in a significant release to atmosphere (via the ‘breathing’ vents of *Safestore*), but it could affect the leachate rates if more graphite surfaces (fragments) became available to water contact.

DISPOSAL OPTIONS FOR GRAPHITE

The ultimate disposal options for the graphite removed from the cores of the dismantled Magnox reactors include the graphite being suitably packaged and disposed of to a land repository or deep ocean; treatment to chemically concentrate or mechanically crush the graphite to a smaller volume for packaging and then disposal; or incineration of the graphite accompanied by some atmospheric release of the radioactive content (primarily carbon-14 and tritium).

To some extent the final packaging and disposal will be determined by the means by which the graphite is extracted from the core which, in itself, might be determined by local conditions and the state of the containment structure. There remains some uncertainty about the practicality and applicability of the present NIREX packaging prerequisites now that the final radioactive waste management strategy is unresolved. These factors (means of extraction, processing and packaging) might be sufficiently important to influence aspects of the final *Safestore* design and its operational time scales.

In the past the UK disposed of graphite wastes via sea dumping but this practice and policy was finally abandoned in 1994 following trades union action in 1982 which had stopped all radioactive sea dumping since that time.

Until quite recently the UK considered incineration to be a viable option, particularly with the graphite being chemically catalysed by pre-treatment with lead-acetate.[11,12] One argument forwarded was that, because of the long half-life of C-14 (5736 years) the impact of short- (incineration) and long-term (repository) did not really differ, and might indeed favour incineration if the efficacy of the various dispersion pathways from a repository were taken into account. Such arguments are believed to continue to find favour in France.

The present proposal for the packaging of graphite (see Nirex, Ref 2) is that it is not to be compacted, nor encapsulated, being placed in its raw state in $4m^3$ mild steel boxes.

---

[1] *The Possibility of Graphite Core Degradation during “Care and Maintenance” and Safestore*”, Wickham A J, et al, AEA/16423595/R/001, April 1996

[2] *The National Radioactive Waste Inventory*, NIREX, 1998

---

[3] *Graphite Core Degradation during "Care and Maintenance" and Safe Storage*", Wickham A J, et al, 1996

[4] *Assessment of the Structural Integrity of the Dungeness A and Sizewell A Cores*, Nuclear Electric TD/NS/REP/0039, Metcalf M, 1992

[5] *Oldbury on Severn, Long Term Safety Review, Graphite Issue 2*, Sadler I, Nuclear Electric, ED/OLA/REP/0054/95, 1995

[6] *Physics of Graphite*, Kelly B Tm, Applied Science Pub 1981

[7] *Stored Energy in the Graphite of Power Producing Reactors,* Phil Trans Series A, V 254, 1962

[8] *Radiolytic Oxidation of Single Crystal Graphite*, Trans Faraday Soc, V 65, 1969

[9] *Graphite Combustion Risk during Decommissioning of the WAGR*, Lomax M, UKAEA, GCRAD(92)93, 1992

[10] *The Effect of Water-Borne Contamination on the Chemical Reactivity of Pile Grade Graphite*, Wilkinson V et al, UKAEA IC+GC-NPCC/MWP/G/P44

[11] *Graphite Disposal Options – A Comparison of Approaches Proposed by UK and Russian Reactor Operators,* Marsden B J Proc Int Conf on Nuclear Decommissioning, IMechE, London 1998

[12] *Assessment of the Management Modes for Graphite from Reactor Decommissioning*, White I F, et al, Nuclear Sc & Technology, EUR 9232 1984