

NUCLEAR GRAPHITE FOR HIGH TEMPERATURE REACTORS

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Abstract:

The cores and reflectors in modern High Temperature Gas Cooled Reactors (HTRs) are constructed from graphite components. There are two main designs; the Pebble Bed design and the Prism design, see Table 1. In both of these designs the graphite not only acts as a moderator, but is also a major structural component that may provide channels for the fuel and coolant gas, channels for control and safety shut off devices and provide thermal and neutron shielding. In addition, graphite components may act as a heat sink or conduction path during reactor trips and transients. During reactor operation, many of the graphite component physical properties are significantly changed by irradiation. These changes lead to the generation of significant internal shrinkage stresses and thermal shut down stresses that could lead to component failure. In addition, if the graphite is irradiated to a very high irradiation dose, irradiation swelling can lead to a rapid reduction in modulus and strength, making the component friable. The irradiation behaviour of graphite is strongly dependent on its virgin microstructure, which is determined by the manufacturing route. Nevertheless, there are available, irradiation data on many obsolete graphites of known microstructures. There is also a well-developed physical understanding of the process of irradiation damage in graphite. This paper proposes a specification for graphite suitable for modern HTRs.

HTR graphite component design and irradiation environment

The details of the HTRs, which have, or are being, been built and operated, are listed in Table 1. A feature of the present designs is that to optimise the power output, an annular core is proposed. This annular core configuration tends to increase the dose to the graphite reflector. The fast neutron flux reduces exponentially with distance into the reflector thus increasing the thermal flux.

During reactor operation, neutron flux and thermal gradients in the graphite components, including the reflector, can lead to component deformations, bowing and the build up of significant shrinkage and thermal stresses.

TABLE 1. HIGH TEMPERATURE GAS COOLED REACTORS

Reactor	Type	MW(t)	MW(e)	Helium Pressure (bar)	Inlet Temperature	Outlet Temperature	Criticality	Shutdown
Dragon	Prism	20		20	350	750	1966	1976
Peach Bottom	Prism	115	40	24	340	715	1967	1974
Fort St, Vrain	Prism	842	330	48	405	780	1974	
HTTR	Prism	30		40	395	950	1999	
GT-MHR	Prism	600		71	288	704		
AVR	Pebble	46	15	11	260	950	1967	1988
THTR	Pebble	750	300	40	250	800	1985	1989
HTR-10	Pebble	10		30	300	900		
PBMR	Pebble	265	110	70	560	900		

In addition the operation of these reactors at high temperature for many years could lead to degradation of the graphite material properties.

The design of the HTR graphite cores must account for these thermal, irradiation conditions. Some of the issues are common to both the prism and pebble bed designs, others are specific to the type of reactor. These design issues are discussed below.

HTR core — prism design

The prism design is best illustrated by the Fort St. Vrain design as described by Neheig, (1972). The main features are the permanent reflector, surrounded by boronated carbon shielding, a replaceable graphite reflector and the hexagonal graphite fuel elements.

Permanent reflector

Although the irradiation fast neutron flux is significantly reduced from the peak value by the time it reaches the permanent reflector, these components must last for the life of the reactor. Therefore the dose the permanent reflector sees may be significant towards the end of life.

The permanent reflector components have to be designed in such a way that the structure remains “gas tight” and that thermal and neutron streaming are minimised. This has to remain the case throughout life and during all thermal and pressure transients, for both normal and emergency operation.

Most designs use graphite keying systems and dowels to keep the components located together. The columns of graphite bricks are usually free standing as individual columns of blocks. This is important as it avoids gapping due to differential thermal expansion and irradiation growth between columns arising from variations in material properties and irradiation behaviour that would occur in a “brick wall” type bonded system.

It is important to provide side restraints for the columns using systems of springs or garters or other support systems. The first major issue connected with the restraint system is the low coefficient of thermal expansion of graphite compared with steel ($\sim 4 \times 10^{-6} \text{ K}^{-1}$ for graphite compared with $\sim 18 \times 10^{-6} \text{ K}^{-1}$ for steel). The second is the large axial and radial temperature distributions within the graphite components that would cause the columns to bow outwards if no radial restraint were provided. The third is the irradiation shrinkage and growth that also leads to column bowing. As changes in the dimensions of graphite components due to thermal gradients and irradiated-induced dimensional changes cannot be prevented, the result of restraining the distortion leads to column kinking which could lead to gas leakage and thermal and neutron streaming if the design did not take this into account. It was small movements of the reflector and fuel element in Fort St. Vrain that led to redistribution of the coolant flow. These resulted in significant changes in fuel outlet temperatures and steam generator inlet temperatures that in turn caused power fluctuations that prevented the achievement of full power operation. To resolve this problem constraining devices were installed on the upper elements (Brey et al., 1982).

In the prism type of reactor design, the permanent reflector receives much less dose than the replaceable reflector. For this reason lower grade graphites were used in Fort St. Vrain and in the HTTR. However, care must be taken in selecting these lower grade graphites as the higher levels of impurities may lead to an increase in decommissioning costs, as discussed later.

Replaceable reflector

The replaceable reflector is normally constructed from blocks of graphite of similar dimensions to the fuel elements. The grade of graphite used in Fort St. Vrain was of a lower grade than that of the fuel. The side replaceable reflector elements were solid, but the upper and lower replaceable reflector had coolant holes and control rod holes. As these components have large flux, and possibly large temperature gradients, across their width and length it is possible that they will become significantly distorted and bowed after several years of operation. This may require prompt replacements, as it was bowing of the replaceable reflector and delaying its replacement that caused problems with fuel removal in the final years of the DRAGON project.

In deciding on the graphite grade for the replaceable reflector, it is important not only to consider the implications for activation of the graphite, but also to consider the cost of obtaining irradiation data to sufficient dose for use in life extension and safety cases. This exercise may be so costly that use of good quality fuel element graphite for the replaceable reflector may be a cheaper option in the long term.

For early permanent moderator prism reactor designs (no longer considered) Blackstone (1969) gives a peak dose of 200×10^{20} n/cm² EDND for a 30 year life time at temperatures between 450-800°C. Clearly a reflector element changed once during its lifetime could only see one quarter of this dose on the side against the fuel.

Fuel elements

There have been various fuel element designs for prism fuel. The behaviour of the individual fuel particles and the compact material in which they are encased are not considered here. However, it should be noted that although the graphite technology associated with the particles and compact is related to that of the main moderator graphite there are differences as non-graphitised materials and possibly natural graphites are used in these fuel items.

The graphite fuel moderator blocks remain in the reactor for much shorter periods than the reflector. However the temperature and flux that they see is more onerous. The blocks may contain passages for the coolant gas and the control rods, as well as holes for the fuel elements and possible burnable poisons.

The large temperature and flux gradients between holes can give rise to significant shrinkage and thermal stresses leading to cracking. For this reason the design and through-life stressing of the graphite prism fuel blocks is important. Three –dimensional assessments are required due to possible end effects in the length of the blocks.

Table 2 gives the doses and temperatures quoted for the lifetime of HTR prism fuel taken from various sources.

Upper and lower graphite structures

Fort St. Vrain is supported on keyed support blocks and post structures in the lower gas outlet plenum. However modifications were suggested for later designs to better resist seismic loading and thermal strains (Peinado, 1982). Above the core there is an upper plenum for the inlet gas, however there is no graphite lining for this structure.

TABLE 2. DOSES AND TEMPERATURES FOR HTR PRISM FUEL

Reactor	Temperature Range °C	Dose $\times 10^{20}$ n/cm ² EDND
UK Mark II		
Fort St. Vrain Maximum [#]	1150	53.6
Fort St. Vrain Median [#]	700 – 950	16.75
Peach Bottom ^{***}	400-800	30-40 (max)
NP-MHTGR [*]	200 and 1300	30
HTTR ^{**}	800-1000	10

* Graphite data requirement for the whole of the reactor.

** Ishihara et al (1998)

*** Everett et al. (1969)

Nehrig et al. (1972)

HTR – Pebble bed design

A typical design of a pebble bed reactor is described by Lohnert and Reutler (1982).

Side reflector

In the pebble bed design the main graphite component to consider from a life-time point of view is the side reflector only. In some earlier designs, as in AVR, there were also graphite “noses” which protruded into the core. The purpose of these noses was to house the control rods, allowing them to be positioned some distance into the core. These noses are no longer a feature of the latest pebble bed designs which have annular core configurations. One of the reasons for the removal of the noses from the design was the high dose environment that they would operate in would probably lead to the need to replace them several times during the reactor life.

The replacement of the reflector in a pebble bed design would be a major undertaking. However, this is being considered in some of the latest designs. The following information is reported on the lifetime dose for pebble bed type cores:

1. From the Dragon programme 140.0×10^{20} n/cm² EDND with a peak temperature of 900°C for a 30 year life (Blackstone, 1969)
2. For the AVR reflector 50×10^{20} n/cm² EDND at 650°C, 12×10^{20} n/cm² EDND at 1000°C (Haag et al, 1986)
3. For the later designs of German pebble bed reactors $300-400 \times 10^{20}$ n/cm² EDND at 730-880°C (Schmidt, 1979).

In AVR the coolant flow was upwards with the return flow passing outside the reflector blocks as was the case in the THTR, where the flow was downwards. However, in the concept for the modular HTR (Lohnert and Reutler, 1982) very large blocks of graphite are used for the reflector, containing holes for the return coolant flow, control rods and small absorber spheres (KLAk-system). The control rod holes are very close to the core boundary where the flux and temperature gradients are high. This can lead to unacceptable shrinkage and stresses at these holes. Various ways have been investigated of overcoming this problem, including slitting the hole, through to the core, to avoid component failure (Schmidt, 1979). This may necessitate the use of graphite sleeves to prevent bypass of the control rod coolant gas. In

addition graphite sleeves may be used in the return gas holes to prevent large temperature gradients and excessive cooling of the large graphite reflector blocks. Another feature of the blocks in the pebble bed design is saucer shaped indentations in the reactor side face of the lower core. These indentations act as “disturbances” to prevent bridging of the outer layers of fuel that may cause these layers to stay longer in the reactor than would be desirable.

The blocks are joined together by a system of graphite dowels and keys and have similar restraint problems related to thermal expansion and dimensional change as discussed for the prism reactor core as discussed above.

The high dose at the surface of the reflector block may cause some of the graphite to be irradiated through shrinkage and “turn-around” until the swelling is much greater than the original volume. At this stage the graphite structure will have started to disintegrate and it is possible that some of the graphite will become so friable that the fuel balls will rub the surface away. Several solutions have looked into solving this problem including sacrificial layers. Another way may be to return to the single zone core with noses and replace the noses at regular intervals.

Lower core, inlet and outlet plenum

The present modular pebble bed designs have both the inlet and outlet ducts at the bottom of the reactor pressure vessel. This arrangement avoids any chimney effect in the extremely unlikely event that the inlet/outlet duct should shear and allow air ingress.

Although the irradiation dose is low at the bottom of the core, there may be large temperature gradients that have to be accounted for. In addition there are gas outlet holes, or slots, that may be subject to compressive loading from the pebbles.

Deeper into the lower core structure the inlet and outlet plenum may pose design problems related to the temperature differences between the inlet and outlet gas. In addition the many paths through the graphite taken up by the inlet and outlet holes, the KLAK systems and the control rod holes can also lead to design problems.

Reflector roof and upper inlet plenum

The irradiation dose to the upper structure is low, however there may be stressing problems related to temperature gradients.

The present pebble bed designs use graphite components for the roof structure and the upper plenum. Roof designs have been either cantilevers as in the HTR module or structures hung from upper structures by metal hangers as in THTR. Both designs have advantages and disadvantages related to component reliability, connectivity with the side reflector and the many holes required for the inlet gas and the control mechanisms.

Graphite manufacture

The starting point in production of graphite is the selection of a suitable coke. These cokes are produced as by-products from the petroleum or coal industry or from naturally occurring pitch sources. These cokes vary considerably in their structure, size and purity.

After production the cokes are broken up and calcined at temperatures between 900-1300°C to drive off volatile material and reduce the amount of shrinkage in the later processes. The calcined cokes are then crushed, milled and graded before being supplied to the graphite manufacturer. It is the choice of the particular coke size, purity and structure that decides the virgin and irradiated properties of the final product.

A suitable blend of coke grades are then mixed with a binder, usually a coal tar pitch. In addition a crushed graphite flour may be added. The coke particles are often referred to as filler particles.

The mixture is then formed into blocks often referred to as the “green article”. Various methods of forming are used and the method chosen has an influence on the properties of the final product. The methods are discussed below:

1. The most common method of production is by extrusion. In this method the mixture is forced through a die under pressure. This method can be used to produce blocks of various sections and of reasonably long lengths. Blocks of the order of 500 mm square by 3600 mm long can be produced in this way. It is important that the extrusion pressure and rate is carefully controlled in order to maintain the desired quality. Graphites produced in this way have anisotropic material properties due to alignment of the filler particle grain, however it is possible to produce reasonably isotropic using this method.
2. Moulding or pressing. This method is used to produce a very isotropic product. The blocks are moulded or pressed from one or two directions at the same time. The AGR graphite moderator blocks were produced using this technique.
3. Iso-static moulding is a more sophisticated method in which the coke and binder mixture is contained in a rubber bag and external pressure applied to give a uniform pressure from all sides.
4. Finally there is vibration moulding. In this method the graphite mixture is placed in a mould, which is vibrated to compact the mixture. Next the graphite mixture is pressed from one side and vibrated again whilst under load.

There are other variations on these methods.

Having formed the ‘green article’, which is reasonably soft, it is rapidly cooled by immersion in water. The green article is then baked at a temperature of around 800°C to drive off more volatile material and ‘cure’ the binder. To prevent oxidation, the blocks are encased in a granular packing, usually a coke. This allows for expansion and helps to support the shape of the green article. This is a long process and may take 30-70 days. One difficulty that arises at this stage is that the thermal conductivity of the graphite is very low, ~30 W/m/K and on cooling thermal gradients in the blocks may lead to internal cracking. One method of overcoming this problem is to add crushed scrap graphite to the mix. However, this has implications for the irradiation behaviour of the final product as discussed later.

The baking process will produce gas evolution pores throughout the structure as volatile gases are driven off. Much of this porosity will be open. To increase the density the baked blocks may be impregnated with a pitch under vacuum in an autoclave. This pitch is much less dense than the binder pitch. To help with this process the surface of the block may be broken by rough machining or by grit blasting. This allows the pitch to enter the open porosity more readily. After impregnation the blocks are re-baked for a much shorter period. There may be up to four impregnations used; however the gain in density for each subsequent impregnation is much less. The product can now be regarded as carbon blocks, which can be used as an

insulation material or furnace liner. However for this application they are usually baked at a higher temperature $\sim 1100^{\circ}\text{C}$.

The carbon blocks are now ready for graphitisation. There are two methods of graphitisation commonly used. The original method is to use an Acheson furnace. This is a large open furnace, which may be up to 7 m wide by 20 m long, into which the carbon blocks are stacked and covered in an electrical conducting coke. A large electric current is applied to each end of the bed through water-cooled electrodes and the blocks are taken through a temperature cycle to $\sim 3000^{\circ}\text{C}$. This process can take about 15 days.

Another, more modern, quicker and cheaper method of graphitisation is to stack the carbon blocks in long lines so that they touch. Again the blocks are covered in coke to prevent oxidation, but this time the current is applied directly through the carbon blocks and not through the packing material. This method can only be used for blocks of similar cross-sections.

During this graphitisation period the graphite crystals are formed and the material becomes much softer and more easily machined. The electrical and thermal conductivity dramatically improve and many more impurities are driven off.

If a more pure product is required the graphite blocks can be reheated to $\sim 2400^{\circ}\text{C}$ in an Acheson furnace with a halogen gas passed through it. However, this final process could add up to 30% to the cost of the graphite. This process of graphite production is summarised in Fig. 1

Polycrystalline graphite microstructure

As discussed above, the final polycrystalline microstructure is determined by the structure of the coke and the binder phase and also by the manufacture process.

At the crystallite level the graphite has strong hexagonal basal planes with much weaker bonding between the planes. For perfect graphite crystals the 'd', or inter-layer, spacing has been measured to be 3.3539 \AA ($0.33539 \times 10^{-9} \text{ m}$) with an 'a' spacing of 2.46 \AA ($0.246 \times 10^{-9} \text{ m}$). The 'c' spacing is twice that of the 'd' spacing.

The size of the crystallites can be measured by x-ray diffraction and has been found to give values of between 400 and 800 \AA for L_a and L_c in well graphitised material (Reynolds, 1968).

In the 'c' direction the size of the crystallite is limited due to so call "Mrozowski" cracks which are formed during cooling from the graphitisation temperature ($\sim 3000^{\circ}\text{C}$), see Fig. 2. The mechanism, which leads to the formation of these cracks, is due to the large difference in the coefficient of thermal expansion in the two crystallographic directions ($\alpha_a \approx 26.5 \times 10^{-6} \text{ K}^{-1}$, $\alpha_c \approx -1.5 \times 10^{-6} \text{ K}^{-1}$). At a temperature of around 1800°C the structure hardens and the much larger shrinkage in the 'c' direction coupled with the restraining affect of the rest of the structure leads to horizontal cracking in the basal planes. Various estimates of the size of Mrozowski cracks have been given, however, in practice there are probably cracks of a variety of widths ranging from less than 250 \AA upwards.

It is this cracked structure that gives graphite its good thermal shock resistance, allowing large crystal expansion in the 'c' direction without leading to inter-crystalline cracking. These

cracks also provide accommodation spaces that can be taken up by irradiation-induced crystal growth and play an important role in determining component property changes in reactor.

Beyond this scale, the cracked crystallite structures are jointed together and follow the general shape of the coke particle. There are also many larger cracks and fissures which also tend to follow the coke particle shape. Examples of this are illustrated in Figs 3. It can be envisaged that the general alignments of the 'a' axis is with the "flow" of the coke particle with the 'c' axis perpendicular to this direction. It can also be envisaged that the shape, size, distribution and orientation of the coke particles will strongly influence the material property of the final graphite component.

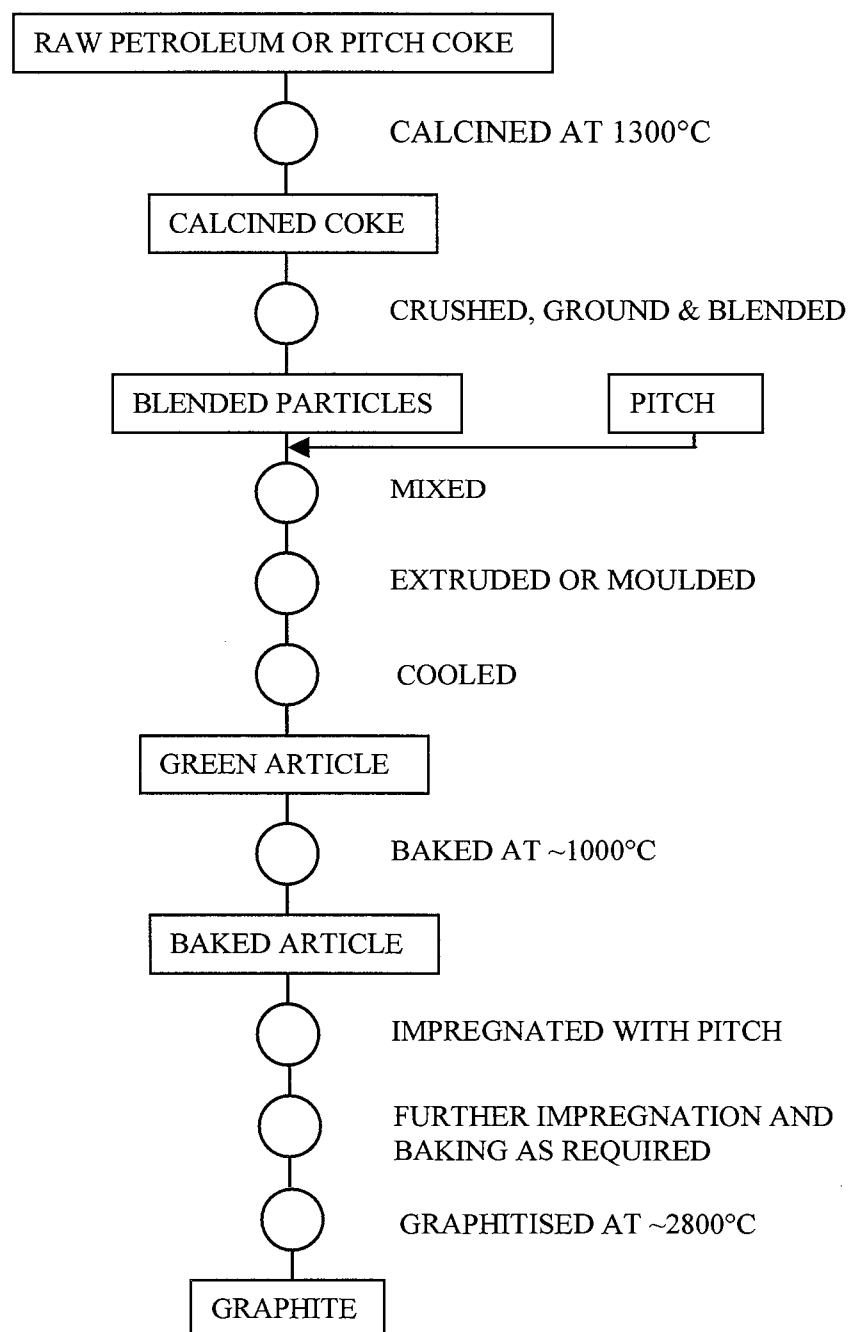


FIG. 1. Graphite manufacturing process.

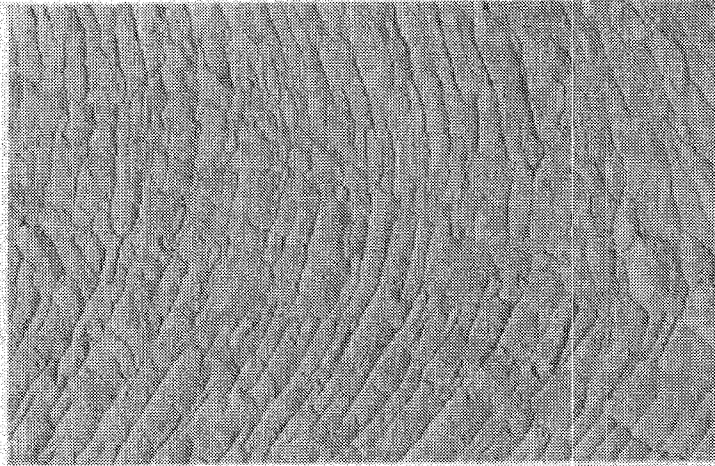
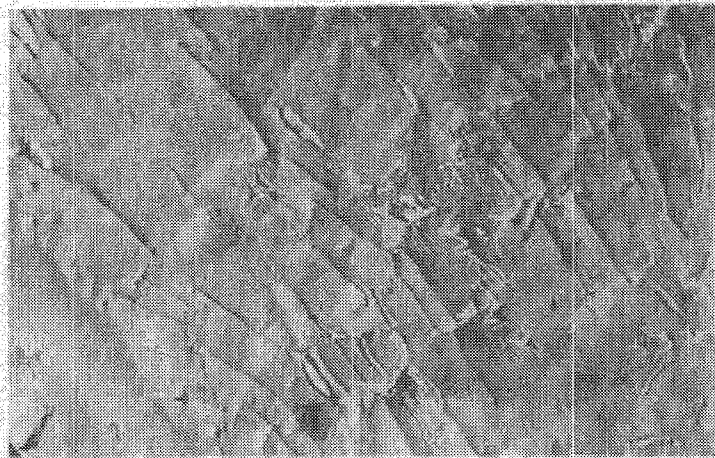
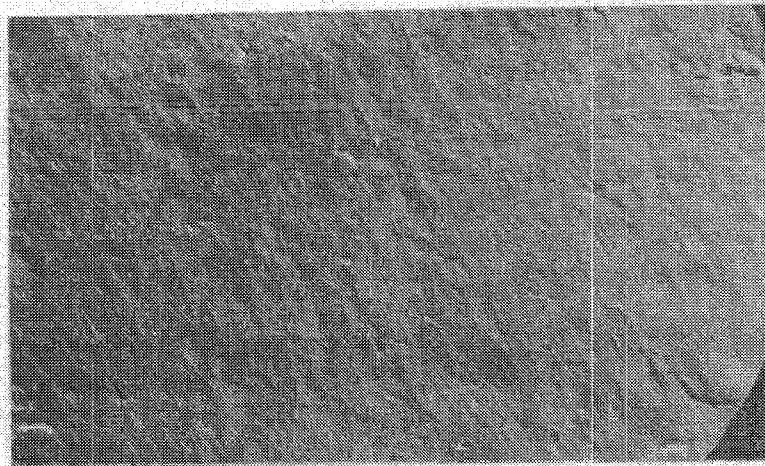


PLATE 7. Electron micrographs of reactor graphite by the replica method
(Magnification $\times 5,000$) (After Thrower and Reynolds, 1963)
(a) Unirradiated.



(b) Irradiated at 200°C to a dose $\gamma_D = 2.7 \times 10^{20}$.



(c) Irradiated at 200°C to a dose $\gamma_D = 25 \times 10^{20}$.

FIG. 2. Microcracks in the graphite crystallite structure.

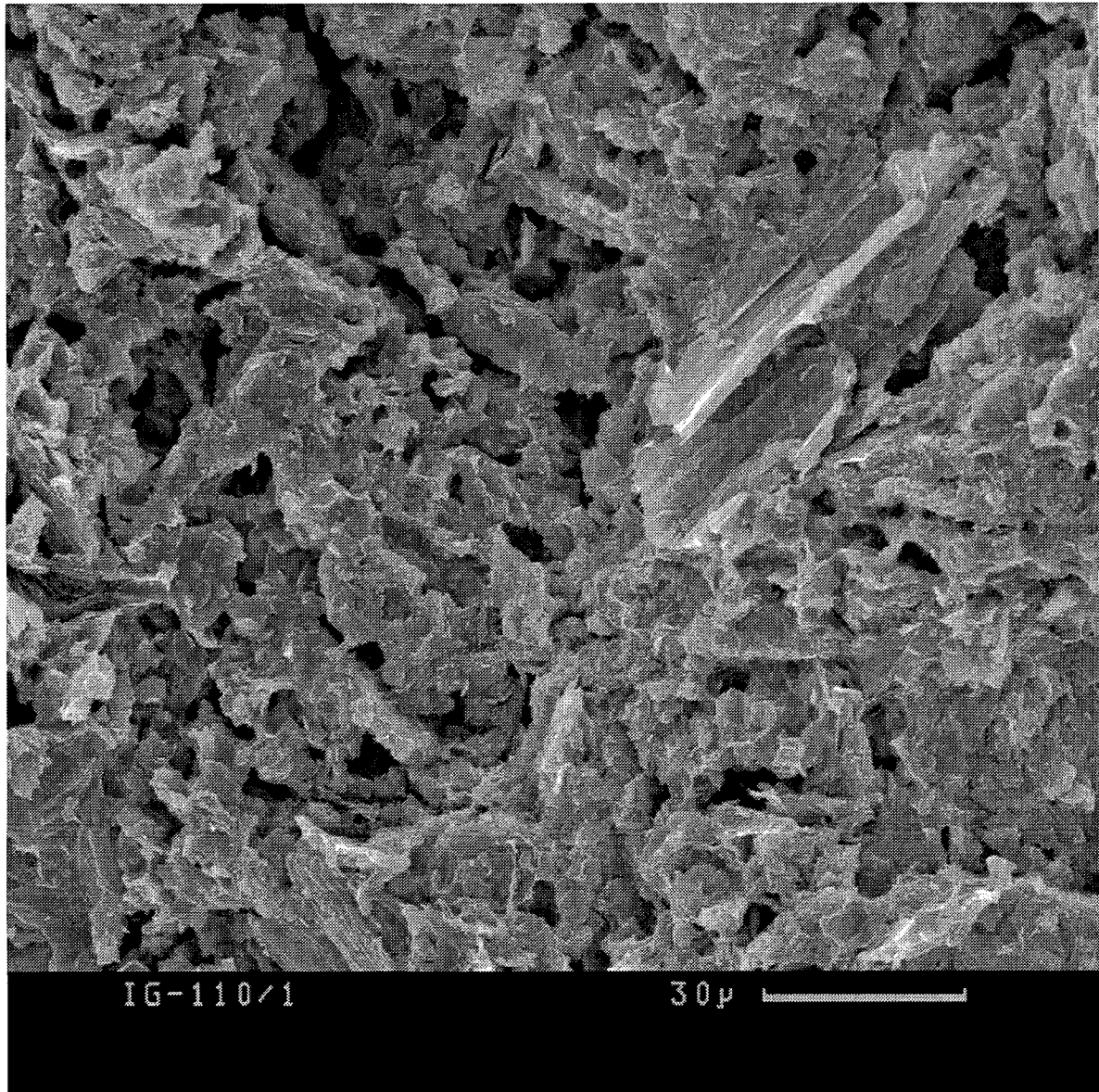


FIG. 3. Micrograph of pitchcoke graphite.

During manufacture the graphite coke particles may have been ground and blended to give a uniform mix. They may be lenticular or needle like in shape or in some special cases spherical. In the UK the Pile Grade A graphite, used in the Magnox reactors was manufactured from a needle coke. Graphite produced from these needle type coke particles is very anisotropic with an anisotropy ratio in the region of 2. The most famous of the graphites with spherical structures is Gilsocarbon graphite which as manufactured from naturally occurring pitch found in remote parts of Utah in the USA. These coke particles formed spheroids with the 'c' crystallographic direction lying mainly in the radial direction and the 'a' crystallographic direction mainly in the hoop. Graphites formed from Gilsocarbon coke had semi-isotropic properties.

Some modern graphites are manufactured from finely ground coke particles, this also can produce semi-isotropic properties.

The coke particles are usually bound together using a pitch binder. The binder itself will probably be mixed with a "flour" consisting of finely ground coke and possibly scrap

graphite. The baked structure may have also been impregnated once or twice to increase the overall density. Features that are of interest are the randomly ordered, well-structured, small particles and the large gas evolution pores.

The orientation of the coke particles in the bulk product is strongly influenced by the forming process. Many types of graphite are manufactured by extrusion which tends to align the coke particles. Other processes described above that can give more isotropic structures are compression moulding and isostatic moulding.

Graphite requirement specification

Density

For a given amount of output, the higher the density of the moderator, the smaller the volume of the core, thus a high-density graphite is desirable. The theoretical density of graphite crystals is 2.265 g/cm^3 , however polycrystalline graphites have a much lower density due to inter-crystalline porosity. Early nuclear graphites had typical densities of 1.6 g/cm^3 , the second generation of graphite moderated reactors used graphites with densities around 1.72 g/cm^3 and the third generation 1.82 g/cm^3 . Modern nuclear graphites have not improved over this latter figure.

The importance of crystallinity

In theory, from a nuclear point of view, any carbon could be used as a moderator if it could be packed to the required density in the required nuclear configuration. However, the availability, and structural properties, of relatively pure artificial graphite led to its use as a moderator in the first graphite moderated nuclear reactors.

These artificial graphites were essentially electrodes used in the steel industry and their purity relied on the choice of raw materials (filler coke and binder) and the manufacturing process and heat treatments to drive off unwanted volatile impurities.

The method used by the graphite manufacturers to determine the degree of crystallinity (or graphitisation) is to measure the electrical resistivity. At room temperature the electrical resistivity can be directly related to the thermal conductivity by the Wiedemann Franz law:

$$\text{Thermal conductivity (kcal/m.h.}^\circ\text{C)} = \frac{0.1116 \times 10^6}{\text{Resistivity}(\mu\Omega\text{cm})}$$

The importance of good crystallinity to the nuclear industry is that it gives an indication of:

- a) a high virgin thermal conductivity
- b) lower crystallite irradiation induced growth than would be the case in poorly graphitised material
- c) lower irradiation induced material property changes (this follows from above)
- d) high purity levels
- e) good machinability.

Nuclear graphite should have a thermal conductivity of around 140 W/m/K , when measured at room temperature.

Dimensional stability

In addition to good crystallinity, polycrystalline graphite components have another requirement to ensure dimensional stability. This is related to the way the randomly orientated crystals in a porous material interact as they grow and shrink with irradiation. Simmons (1965) demonstrated that there was an empirical relationship between dimensional change rate and the Coefficient of Thermal Expansion (CTE) (The higher the coefficient of expansion the less the dimensional change rate). He also derived a theoretical relationship between dimensional change rate and the coefficient of thermal expansion (Marsden, 1998). However this relationship breaks down at comparatively low doses.

It was also found that extruded coarse grain graphites that have anisotropic material properties tended to have lower CTEs than more isotropic graphites. However, it was also observed that some isotropic graphite with very high CTEs expanded with irradiation.

From experience it is desirable to choose a graphite with a coefficient of expansion between 4.0 and $5.5 \times 10^{-6} \text{ K}^{-1}$ measured over the range $20\text{-}120^\circ\text{C}$.

Air reactivity (thermal oxidation in air)

In the unlikely event of an incident involving air ingress, it is important that the air reactivity, that is the rate at which graphite can oxidise in air, is as low as possible. Irradiation increases the air reactivity rate in graphite, however this effect is minimal compared to the increase in air reactivity caused by catalytic impurities.

For the AGRs the mean reactivity in air of samples was specified as not exceeding $3 \times 10^{-6} \text{ g/g/k}$ at 400°C (Hutcheon and Thorne, 1965).

Absorption cross-section

For the particular HTR design the nuclear physics considerations will lead to the values of absorption cross sections required. However, historically there has been no systematic effort put into determining a specification for the absorption cross-section in graphite.

The early plutonium production reactors built in the UK used various graphites with nuclear absorption cross-sections of between 4.7 and 5.1 mbarn. The later military and civil carbon dioxide cooled reactors used Pile Grade A (PGA) graphite with an absorption cross-section of 4.0 mbarn. For this reason an upper limit for boron of 0.2 ppm was specified. The method used to measure the absorption cross section was to place graphite samples, or whole blocks in some cases, in a test reactor such as GLEEP in the UK.

Later when the AGRs were designed it was recognised that the major impurity atom contributing to the absorption cross section in graphite was ^{10}B which burnt up rapidly early reactor life. For these later reactors the absorption cross-section was calculated from the chemical inventory, however the value arrived at depended on what impurities were measured and the choice of measurements was somewhat arbitrary.

As previously discussed it is clear that in the past no systematic effort has been made to define a specification related to the impurity levels related to absorption cross-section. The author considers that a list of elements with significant absorption cross-sections should be identified

as part of a graphite specification. The graphite manufacturer could then be asked to analyse their product for these elements.

Impurities related to operational and decommissioning problems

The radioactivity associated with graphite components arises from initial impurities and from subsequent contamination within the reactor circuit.

Probably the most important isotopes related to initial contamination will be ^{60}Co , ^{154}Eu , ^3H , ^{36}Cl , ^{41}Ca and ^{14}C , however this is not an exhaustive list and a more rigorous approach is required as discussed below.

An IAEA TechDoc (Marsden, 2001) is being prepared at present on the subject of decommissioning graphite reactors which gives much more detail concerning the problems related to disposal of nuclear graphite.

Releases during operation

In the UK, graphite reactors such as the AGRs and Magnox have to monitor the release of certain isotopes. Some of these radionuclii, such as ^{36}Cl , ^{14}C and ^3H , may be related to the release of products from the graphite core. Again a definitive list of undesirable isotopes, which may be released from graphite, is required, taking account of regulations in the country where the reactor is to be constructed. The likely origins of these radionuclii can then be determined to give a list of undesirable impurities from an operational point of view. Information will also be required as to how easily these radionuclii can be released from the graphite structure during operation.

Decommissioning

Similarly a list of the most undesirable radionuclii with regard to the long-term risks associated with waste disposal can be obtained from the appropriate authorities, such as NIREX in the UK. Then the route that can produce these radionuclii can be determined to give a comprehensive list of undesirable elements from a decommissioning point of view.

Strength

In a modern reactor, the strength of the graphite is important, as it may be subjected to shrinkage and thermal stresses, as well as restraint loads and possible seismic impact loads during the life of the reactor. Irradiation modifies graphite strength, as does thermal and radiolytic oxidation.

Graphite is stronger in bend than tension and stronger in compression than bend. As with many brittle polycrystalline materials, the failure strength depends on the component geometry, loading configuration and component size. Unfortunately at present there is no satisfactory failure model for unirradiated, or irradiated, graphite which makes it difficult to predict the behaviour of graphite components in the future.

Proposed Specification for HTR Graphite

1. The graphite should be reasonably dense $\sim 1.8 \text{ g/cm}^3$.

2. It should be well graphitised as indicated by a thermal conductivity of ~145 W/m/K measured at room temperature.
3. It should have a low absorption cross-section, between 4 and 5 mbarns. (This can be calculated from knowledge of the chemical impurities).
4. Impurities that could possibly lead to operational problems and high decommissioning costs must be kept to a minimum.
5. The graphite must have a high irradiation dimensional stability. This is indicated by a relatively high CTE(20-120°C) between 4.0 and $5.5 \times 10^{-6} \text{ K}^{-1}$.
6. The irradiation time, over the irradiation temperature of interest, for the graphite to return to its original volume should be as long as possible. (In the long term a Material Test Reactor (MTR) programme can only confirm this.)
7. The graphite must have a moderately high strength (A tensile strength of about ~20 MPa).
8. The air (moisture) reactivity should be measured to ensure that the rates are acceptable (0.2 to 0.01 mg/g-h).
9. A suggested list of chemical impurities that should be minimised is given in Table 6. This list is based on past experience and should be reviewed in the light of local requirements for decommissioning and operation.

TABLE 6. GRAPHITE IMPURITIES THAT ARE CONSIDERED TO BE INCOMPATIBLE FOR REACTOR OPERATION AND FINAL DECOMMISSIONING AND DISPOSAL

Element	Symbol	Element	Symbol
Aluminium	Al	Mercury	Hg
Antimony	Sb	Manganese	Mn
Arsenic	As	Molybdenum	Mo
Beryllium	Be	Nickel	Ni
Barium	Ba	Chlorine	Cl
Boron	B	Potassium	K
Bismuth	Bi	Phosphorous	Pb
Cadmium	Cd	Platinum	Pt
Caesium	Cs	Selenium	Se
Calcium	Ca	Samarium	Sm
Chromium	Cr	Silver	Ag
Cobalt	Co	Silicon	Si
Copper	Cu	Sodium	Na
Gold	Au	Tantalum	Ti
Indium	In	Tin	Sn
Hafnium	Hf	Sulphur	S
Lead	Pb	Titanium	Ti
Dysprosium	Dy	Tungsten	W
Europium	Eu	Vanadium	V
Iron	Fe	Zinc	Zn
Gadolinium	Gd	Strontium	Sr
Lithium	Li	Halogens	
Magnesium	Mg	Rare earth metals	

Conclusions

Unirradiated and irradiated graphite material properties depend strongly on the choice of the raw materials and the manufacturing process. Suitable graphite for modern HTRs can be designed based on the choice of coke, binder and manufacturing technique. However a compromise is always necessary. A suggested specification for HTR graphite is given.

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