

High Temperature Gas-cooled Reactor Core Design Future Material Consideration

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To remain competitive, future HTGRs (High Temperature Gas-cooled Reactors) will require a core and reflector design that is capable of operating reliably at very high temperatures for extended lifetimes (~40 years) with the minimum generation of radioactive waste. The present core designs are based on the use of high quality, semi-isotropic nuclear grade polycrystalline graphites. Whilst these materials have proved to be robust and reliable when used in prototype and early commercial HTGR designs, nuclear graphites presently available are likely to be pushed to the limit in future applications. The main limitations of current nuclear graphites can be directly related to fast neutron damage at the microscopic and macroscopic level. When using the currently available graphites, the HTGR designer has to consider component replacement earlier than would be desirable. Therefore, for future systems the prospective HTGR designer would like to know if the present graphite types could be improved upon. Are other materials available that may replace conventional graphite? Can innovative design be used to address these difficulties? If they do have to replace core components, can the amount of radioactive waste be reduced? This paper discusses HTGR core design from a materials aspect, looking forward to where effort may be required when addressing future HTGR core material requirements.

KEYWORDS: *graphite, HTGR, HTR, design*

I. Introduction

The new designs for High Temperature Gas-cooled Reactors (HTGR) require graphite components that will withstand higher temperatures to higher fast neutron doses that have been previously achieved. In addition the amount of radioactive graphite waste produced during operation must be kept to a minimum. The irradiation-induced changes of importance in nuclear graphite are dimensional changes and changes to the Coefficient of Thermal Expansion (CTE), Young's modulus, strength, thermal conductivity and irradiation creep. All of these property changes are a function of irradiation dose and temperature. These changes result in a life limit for HTGR graphite components determined by one of two factors:

- Generation of internal and thermal stresses leading to component failure.
- High dose degeneration of the microstructure and loss of its cohesive strength.

The first factor can be addressed, to some degree, through design of the graphite components and is not discussed in this paper. However, the second factor is related to the initial microstructure of the particular graphite used.

Graphite is produced from cokes and pitches that are bi-products of either the coal or oil industry. The choice of

coke and the manufacturing technique determine the irradiation behaviour. No two graphite types behave exactly the same with irradiation, although semi-isotropic graphites obey similar basic trends with dose and temperature. It has been traditional to obtain irradiation empirical data for particular graphites through a Material Test Reactor (MTR) programme. However, as coke supplies change from time to time there is a need for a new MTR programme every time a new graphite is chosen for a nuclear application. This may take up to eight years at considerable cost and thus, such programmes are not practical within the constraint of the present economics related to nuclear power. Therefore, for future HTGR designs there is a need for a predictive mechanistic understanding of why graphite behaves as it does with irradiation. This paper discusses how to improve the understanding of nuclear graphite behaviour and how this understanding may be used to envelope new graphitic materials for HTGRs.

II. Graphite Requirements for Future HTGRs

When selecting graphite for the AGRs in the 1960s, it was realised that semi-isotropic graphite behaves much better than anisotropic graphite and that graphite with a relatively high CTE exhibited lower dimensional change than graphite with low CTE. Thus, this report only concentrates on high CTE, semi-isotropic graphite.

The relationship between typical irradiation-induced changes in graphite is illustrated in **Fig. 1**. The rapid growth after the dimensions have returned to the original volume leads to rapid degeneration of the properties, particularly

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strength and modulus. The return to original volume can therefore be seen as an indication that the graphite is approaching its useful life limit. The higher the irradiation temperature the further to the left these curves move along the dose axis and the shorter the life of the graphite. At a irradiation temperature of around 1000°C, the rate of initial shrinkage increases, probably due to thermally induced micro-crack closure. Thus in an ideal graphite, the shrinkage behaviour should be minimised and the point at which the change in volume returns to zero must be as far to the right in dose as possible, as defined by Fig. 1. As the graphite is to act as a moderator it should be reasonably dense ($>1.7 \text{ g.cm}^{-3}$) and have a low absorption cross-section ($\sim 4 \text{ mbarn}$); the latter can be calculated from knowledge of the chemical impurities. It should also be well graphitised as indicated by a thermal conductivity of $>100 \text{ W.m}^{-1}.\text{K}^{-1}$ when measured at room temperature. Impurities that could lead to operational problems and high decommissioning costs must be kept to a minimum. The graphite should have moderate strength (a tensile strength of $\sim 20 \text{ MPa}$). The air (moisture) reactivity should be measured to ensure that the rates are acceptable (approximately $20 \text{ mg.g}^{-1}.\text{h}^{-1}$).

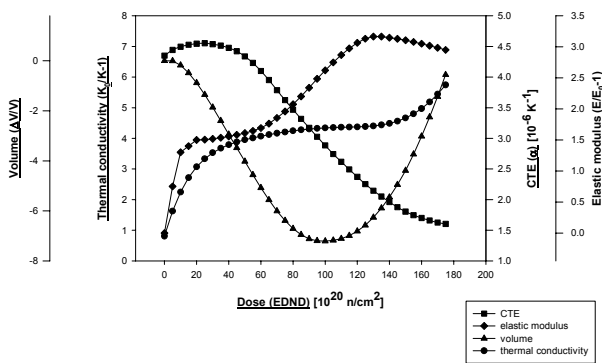


Fig. 1 Typical irradiation induced changes in a nuclear graphite

III. Property Change Understanding Through Microstructural Change Understanding

Nuclear graphite is a polycrystalline material that is extremely pure (impurities are measured in ppm). It is formed from coke particles mixed with a binder before baking ($\sim 800^\circ\text{C}$, 20-72 days) and graphitising ($\sim 3000^\circ\text{C}$, ~ 15 days). Within the coke particles there is well graphitised material with a well-developed graphitic structure (**Fig. 2**) separated by long cracks of various widths, parallel to the crystal basal planes. These cracks, along with other porosity associated with gas evolution during manufacture, mean that artificial graphite has a density of between 1.6 and 1.85 g/cm^3 as opposed to a theoretical density of 2.26 g/cm^3 . Stress / strain deformation of the crystal structure is through basal plane sliding, dominated by the movement of glissile dislocations. Fast neutron irradiation knocks atoms out of the crystal lattice in a cascade, leading to the formation of interstitial and vacancy loops parallel to the basal planes.

This leads to crystal growth in the 'c' direction and shrinkage in the 'a' direction. When polycrystalline graphite is irradiated, porosity, particularly the cracking parallel to the basal planes, causes the 'a' shrinkage to dominate, until most of the basal aligned cracks have closed, hence the 'c' growth is observed. Other crystal changes, such as dislocation pinning, and the increase in irradiation-induced defects lead to an increase in crystal shear modulus and a decrease in thermal conductivity.

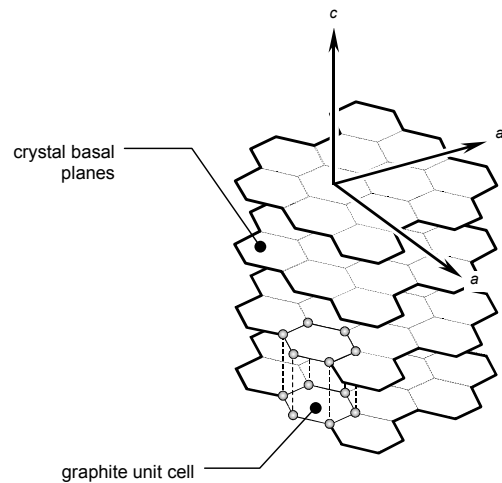


Fig. 2 Graphite hexagonal structure

Thus, to gain an understanding of the irradiation changes and to provide tools to develop a graphite for future HTGRs, an understanding of the relationships between graphite crystal dimensional change, graphite crystal property changes (modulus, CTE and thermal conductivity) as a function of irradiation dose and temperature is required. There is a need for not only the dimensional and property change measurements, but for electron microscopy studies to accompany them. Although some data does exist for the temperatures of interest for the new reactors, the database is poor. By using good crystal data it should be possible to derive validated, semi-empirical equations for irradiated crystal behaviour throughout the required dose and temperature range. From previous studies it is clear that the best representation for artificial graphite simulated behaviour is Highly Orientated Pyrolytic Graphite (HOPG) heat-treated to the graphitisation temperature ($\sim 3000^\circ\text{C}$).

Having gained this understanding of irradiated crystal behaviour, the next step is to develop an understanding of the relationship between the polycrystalline microstructure and the crystal property changes. With respect to dimensional change, CTE and modulus this is probably best accomplished by examining the microstructure and making use of finite element techniques.

From the two such stages described above, it should be possible to determine the ideal coke structure to give an

optimum life for the graphite. Research would then be needed to produce such cokes. Alternatively, the analysis may point to other graphitic structures that could be used to produce alternative products.

IV. Need for the Implementation of a Graphite MTR Programme

From the discussion above it is clear that a graphite MTR programme is required, which instead of focussing on empirical data, focuses on gaining a mechanistic understanding of irradiation behaviour in graphite. This study should be comprehensive, systematic and compose of two stages:

- Stage 1 – to gain an understanding of irradiation-induced dimensional and property changes in graphite crystallites over the temperature range relevant to HTGR.
- Stage 2 – to gain an understanding of how these changes influence the polycrystalline dimensional and property changes over the same temperature range.

These programmes should also include the use of microscopy, x-ray tomography and other modern imaging techniques. It is also essential that the work is accompanied by modelling techniques. One such modelling technique is described below.

V. Modelling graphite irradiation behaviour

Numerous authors have examined the relationship between crystal property changes and observed polycrystalline changes, and how the former can be used as the basis of predictions. Simmons¹⁾ derived a set of relationships for dimensional change rate and CTE that associated the macroscopic properties of a polycrystalline graphite to the properties of graphite crystals. A similar set of equations was obtained by Jenkins²⁾ with the exception that the CTEs were related to elastic moduli, orientation and absorption of expansion by voids. Kelly and Brocklehurst³⁾ expanded the Simmons relationship to account for pore generation, and subsequently developed it further^{4,5)}. A modification of the original Simmons relationships was also proposed by the authors⁶⁾.

All of these relationships had used property changes in graphite crystals and polycrystalline graphite to give the required changes of another polycrystalline property. They did not give an indication of what was actually happening to the microstructure. Also, the relationships considered graphite to be single phase, with or without pores. However, nuclear grade graphite is not a single-phase material. As stated previously, it is made from two distinct constituents; filler and binder. Morgan⁷⁾ used a two-phase model to examine the irradiation-induced dimensional changes in graphite. The two phases were defined as ordered graphite crystallites (primary phase) and

non-graphitic carbon (secondary phase); the primary being akin to filler and the secondary to binder. The model also included oriented microcracks caused by cooling in manufacture⁸⁾. However, because of the complexities of such a model, little work was done on it and the single-phase method remained the most popular.

It was not until the advent of powerful computers to use the finite element method that the two-phase approach would be used again. The current method uses the finite element program ABAQUS to model a two-phase graphite from the microscale (coke or filler particle) to the macroscale (bulk graphite).

1. Graphite Filler Model

Using the work by Sutton and Howard⁹⁾ and White¹⁰⁾, an idealised filler model was developed. A quantity of well-crystallised, cracked material (filler) was surrounded by a layer of amorphous material (binder). Internal cracks were added to represent Mrozowski-type cracks or accommodation porosity. **Figure 3** gives an example of such a model, although numerous models were created to give the required distribution of sizes and percentages.

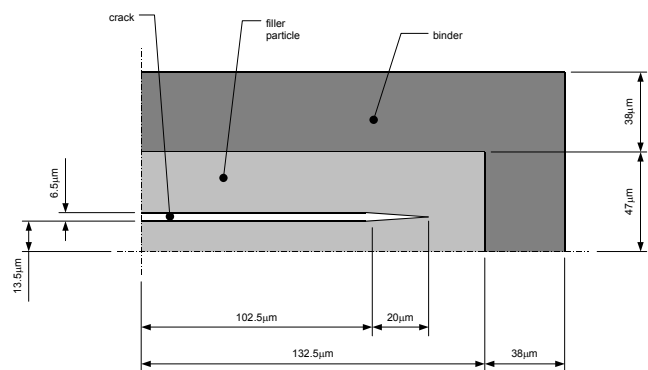


Fig. 3 Graphite filler model

The analysis was conducted using a subroutine facility within ABAQUS called User Material (UMAT) that allowed the definition of a material's constitutive equations. In the case of the filler material these were anisotropic, temperature and irradiation dependent relationships for:

- crystal dimensional change ($\Delta a/a$, $\Delta c/c$)
- crystal CTE and changes in CTE with strain (α_a , α_c)
- crystal elastic modulus (E_a , E_c)
- crystal Poisson's ratio (ν)
- crystal irradiation creep (ϵ_c)

The input data used in these relationships were taken from experiments on HOPG, the closest practical representation of a crystal. Dimensional changes were obtained from Brocklehurst and Kelly¹¹⁾, and the elastic modulus and Poisson's ratio were from Kelly¹²⁾, and Seldin

and Nezbeda¹³⁾ respectively. In the models described in this paper it was assumed that the elastic modulus was independent of dose and temperature. The CTE data was acquired from studies by Bailey and Yates¹⁴⁾ and Morgan¹⁵⁾. In the current models, the effect of irradiation creep is ignored.

The binder material was given dose independent material properties based upon typical values for an isotropic nuclear grade graphite (Gilsocarbon). This material was located around the filler material to represent the restraint imposed by the surrounding filler particles and binder. Without this, the filler particle would only change in accordance with the respective crystal dimensional change and CTE for each direction.

By cycling between the irradiation temperature and room temperature at regular intervals of dose, it was possible to extract the apparent dimensional changes and apparent CTE (Fig. 4). The apparent elastic modulus was determined by loading the filler particle in compression or tension at regular dose intervals (Fig. 4).

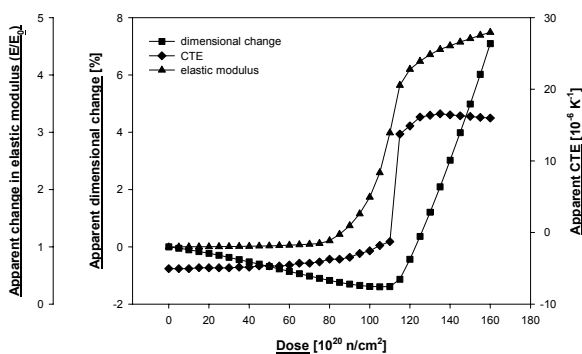


Fig. 4 Filler model apparent property changes ('c' axis)

The results from the filler modelling would then be used as inputs for a polycrystalline model.

2. Polycrystalline Graphite Model

This model comprised of a square mesh of elements (Fig. 5) in which individual elements may represent:

- a filler particle containing cracks, surrounded by binder materials (the filler particle models)
- binder material
- a hole that symbolises inter-crystalline porosity or gas evolution pores.

The location of these elements was randomly generated but the amount and distribution for each could be specified to match the graphite under scrutiny. In an isotropic case such as Gilsocarbon, the amount of filler, binder and porosity was taken from manufacturing information by Nightingale¹⁶⁾. The accommodation porosity was given a

log-normal distribution with the mean value of 5%, set according to Sutton and Howard⁹⁾. The filler particles were given an even distribution of orientations in order to give an isotropic behaviour.

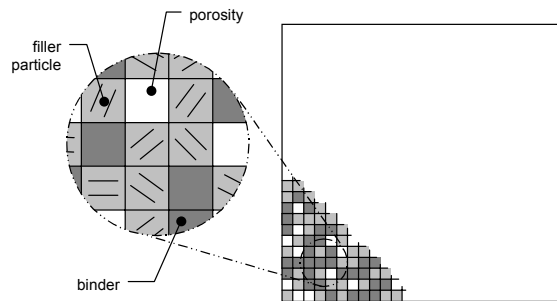


Fig. 5 Polycrystalline graphite model

The polycrystalline model included a simplistic form of cracking. In polycrystalline graphites at high doses, a reduction in elastic modulus has been observed. This can be linked to cracking of the structure¹⁷⁾. Shtrombakh *et al.*¹⁸⁾ identified a mechanism where the filler/binder interface cracks as the dose increases. To account for this in the model, a stress limit was set to the binder material, after which the elastic modulus of the binder material would reduce until it reached a set minimum.

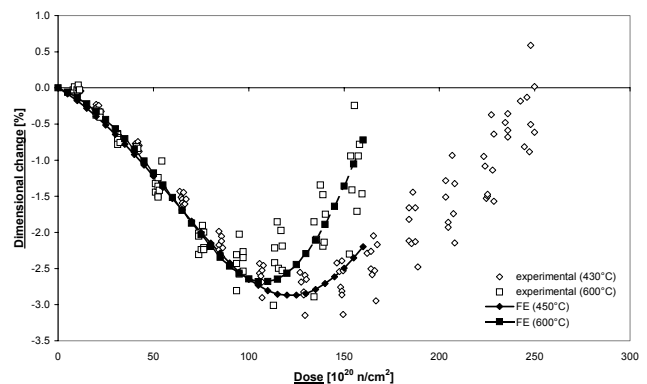


Fig. 6 Polycrystalline dimensional changes

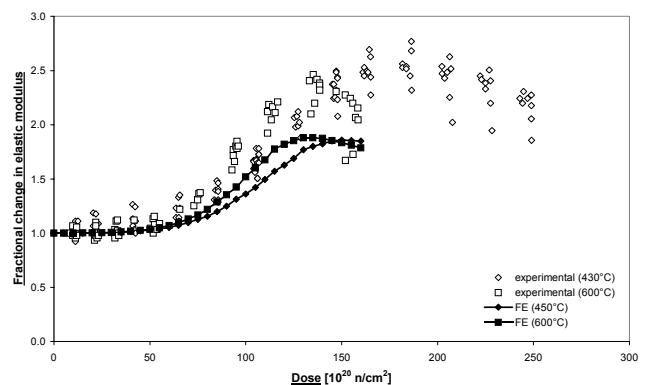


Fig. 7 Polycrystalline elastic modulus changes

The polycrystalline model was loaded as per the filler model and the apparent dimensional changes (**Fig. 6**), elastic modulus (**Fig. 7**) and CTE extracted. However, it should be noted that initial studies showed that the polycrystalline CTE was unrepresentative of the observed behaviour and therefore, emphasis has been placed on dimensional change and elastic modulus.

Comparison of the finite element results with experimental data⁵⁾ showed some striking similarities. At the two irradiation temperatures considered, the dimensional changes demonstrated the same trends and lay within the scatter of the data (Fig. 6). The elastic moduli at the two temperatures also showed good correlation to the experimental data, although again the magnitudes were different (Fig. 7).

The modelling procedure has led to some significant findings. As it was possible to explicitly model the cracks it has been determined that, in these models at least, turnaround and the increase in elastic modulus with dose can be attributed to crack closure. The reduction in elastic modulus has also been associated with microstructural cracking. It has been possible, with some success, to transfer the procedure between two quite different graphites. This is important because if the property changes examined are truly a result of microstructural changes, the procedure should be applicable to any type of graphite. However, to get further understanding and confidence in the procedure it will be necessary to take it to other graphite types (IG110 etc.) and operating conditions (higher temperatures, radiolytic oxidation etc.).

The perceived problem with this method is the scarcity of data. The procedure is dependent upon the input data to the filler model, of which there is limited available. There is also limited polycrystalline data for comparison. In some cases extrapolations and assumptions had to be made.

VI. Issues Surrounding Irradiated Graphite Waste

Nuclear graphite is an extremely pure product. However, trace elements will inevitably exist within the structure. In addition, as graphite is a porous material it is possible that at the end of life it may be contaminated by a limited amount of radio-nuclei carried around the circuit, including some fission products from fuel failures. Limitation of the latter problem is not the graphite manufacturers' concern. However, the former is the concern of the manufacturer and it is essential that trace elements such as cobalt, lithium and chlorine, resulting in ⁶⁰Co, ³H and ³⁶Cl, be kept to a minimum. The latter may be present due to its use in the purification process during manufacture. Carbon-14 is mainly produced in graphite¹⁹⁾ through the presence of nitrogen, and as graphite is a porous material and air is available during the manufacturing process, it is difficult to see how traces of nitrogen can be excluded. Graphite waste in many cases is considered to be intermediate level waste.

Prevention of leaching of the radio-nuclei may be accomplished by resin impregnation or some other treatment, thus making shallow burial disposal an option. Alternatively it may be packaged in a suitable grout and disposed of in a deep geological repository. Another option may be incineration which has the advantage that considerable waste reduction is achieved, but the problem of ¹⁴C release requires addressing if this option is to be considered.

VII. Other Possible Materials

There has been much interest in the use of carbon reinforced carbon composites for nuclear applications, particularly in the field of fusion. However, whilst these products have good thermal properties, their ability to withstand fast neutron damage appears to be limited. To date the study of such material has relied on serendipity and it is the authors' opinion that work in this area require more focus along the lines suggested for graphites in order to determine how such materials can be engineered. However, this does not preclude their present use in HTGR technology in regions where the dose is low, such as for core restraints and supports, where their excellent strength and thermal properties can be taken advantage of.

A possibility is to develop specifically designed coke particles that have an amount of porosity within their structure to give the optimum dimensional change behaviour. Another option may be to develop spherical carbon particles along the concept of the HTGR particle fuel, in which the pyrocarbon layer arrangement is designed to give ideal shrinkage behaviour. Alternatively, fibres or tapes could be developed to form the basic filler material, again with the aim being to create desirable bulk material behaviour.

The modelling described in this paper, in conjunction with an MTR programme, may be of aid in the design and prediction of irradiation behaviour of such materials.

VIII. Conclusions

1. There is a need for a database for pyrolytic graphite behaviour over the HTGR dose and temperature range. The database should include dimensional change, CTE, elastic modulus and thermal conductivity.
2. A method of modelling that may be used to predict the behaviour of polycrystalline graphite is described. This technique may also be used in the design of "new" graphites.
3. The possibility of developing new coke and innovative graphite structures such as spheres, tapes and fibres requires investigation.

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