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Comparison of Material Behavior of Matrix Graphite for HTGR Fuel Elements upon Irradiation: A literature Survey

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1. Introduction

It is well known that the former German HTGR programs have been and the Chinese HTGR programs are being based on the pebble bed core design while the US HTGR programs and the Japanese HTTR program are based on the prismatic core design. The fuel elements for the HTGRs (i.e., spherical fuel element in pebble-bed type core design and fuel compact in prismatic core design) consists of coated fuel particles dispersed and bonded in a closely packed array within a carbonaceous matrix. This matrix is generally made by mixing fully graphitized natural and needle- or pitch-coke originated powders admixed with a binder material (pitch or phenolic resin), The resulting resinated graphite powder mixture, when compacted, may influence a number of material properties as well as its behavior under neutron irradiation during reactor operation. In the fabrication routes of these two different fuel element forms, different consolidation methods are employed; a quasi-isostatic pressing method is generally adopted to make pebbles while fuel compacts are fabricated by uni-axial pressing mode.

In this review, the material behaviors of the matrix graphite under neutron irradiation in these two different fuel elements are compared, on thermal and mechanical properties, in particular. Also discussed are the fabrication process and relevant specifications applied for the respective fuel element designs.

2. Fuel Element Fabrication Processes

The fuel element for an HTGR is manufactured by mixing coated fuel particles with matrix graphite powder and forming them into either pebble type or cylindrical type compacts depending on their use in different HTGR cores.

The basic steps for manufacturing a fuel element include the preparation of the graphite matrix powder, over-coating the fuel particles, mixing the fuel particles with a matrix powder, forming green fuel pebble or green fuel compact, carbonizing, and a final high-temperature heat treatment of the carbonized fuel pebbles or fuel compacts. In Fig. 1 and 2, recently established fuel element fabrication processes are schematically illustrated for the Chinese HTR-10 and the Japanese HTTR, respectively. [1, 2]

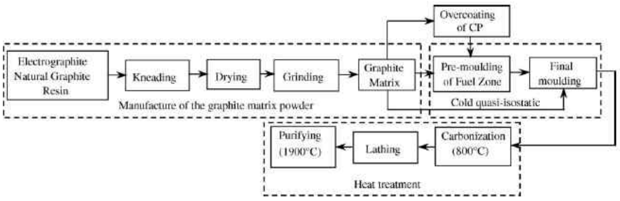

Fig. 1. The fabrication process flow for the Chinese HTR-10 spherical fuel elements

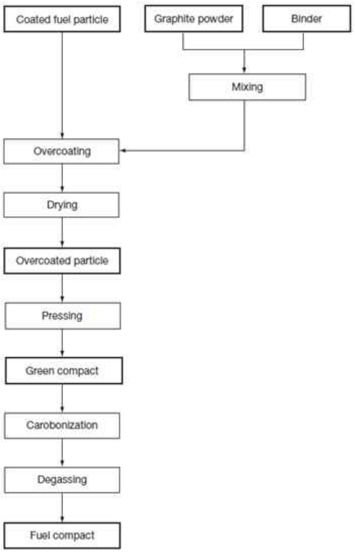

Fig. 2. The fabrication process flow for the Japanese HTTR fuel compacts.

The material properties of the matrix graphite are greatly influenced by the graphite matrix powder preparation process, which is divided into several steps; 1) mixing of natural and artificial graphite powders, 2) admixing and kneading the powder mixture with binder solution and transforming graphite matrix paste into cake, 3) drying graphite matrix cake, 4) milling graphite matrix cake and 5) sieving. In each step, the process parameters are strictly controlled in order to achieve the appropriate properties of the final milled graphite matrix powder for further processing, e.g., mixing with coated fuel particles and pressing the mixture of graphite matrix powder and coated particles. Fig. 3 illustrates the steps involved in the graphite matrix powder preparation process.

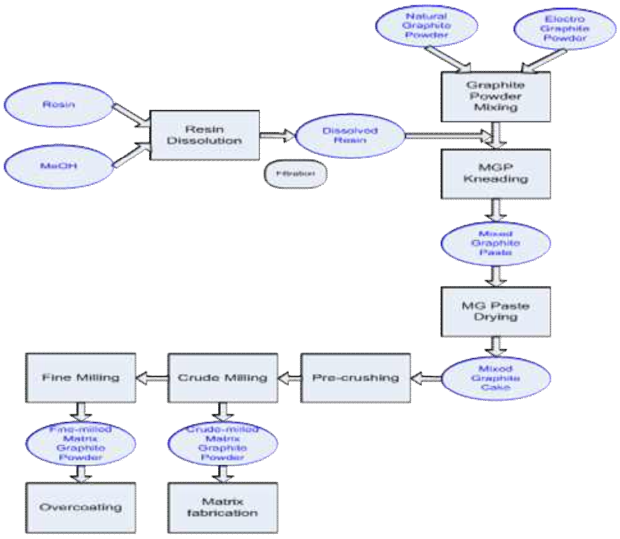

Fig. 3. Detailed process flow of graphite matrix powder preparation.

3. Difference in Functions of Graphite Matrix between Spherical Fuel Element and Fuel Compact

Prerequisites of spherical fuel elements in a pebble-bed type design [3] require that the matrix graphite 1) act as moderator for fission neutrons, 2) provide for heat transfer from the surface of the coated particles to the surface of the fuel element, 3) protect external forces, 4) be resistant against corrosive attack and 5) maintain high dimensional stability during irradiation with fast neurons. The corresponding key properties of the pebble matrix to be kept within specified ranges are, therefore, graphite density, thermal conductivity, mechanical strength, dimensional stability and corrosion resistance.

Meanwhile, as the fuel compacts in a prismatic core design are inserted in graphite fuel blocks, the requirements of the matrix graphite can be summarized as follows [4]:

1) to have a relatively good thermal conductivity so as to minimize compact and cross particle thermal gradients,
2) to provide local mechanical support for the coated particles without risk of damage to the outer coating caused by local thermal or irradiation induced stresses, i.e., bonding to the particle should be relatively weak and matrix cracks should not propagate through particles,

3) to result in a fuel body which will withstand internal thermal and irradiation induced dimensional changes and stresses without disintegration, i.e., it should have good strength, adequate irradiation creep, suitable Young's moduli and coefficient of thermal expansion,
4) the thermal and irradiation induced dimensional changes of the fuel body should be such that interaction with and fracture of the cladding graphite or at the other extreme, the creation of large helium gaps which would result in an undesirable increase in fuel temperatures, are avoided,
5) to act as a sacrificial layer in the event of chemical attack by oxidants in the coolant, should a fuel tube be fractured, i.e., the matrix should minimize the physical detachment and chemical attack on particles,
6) ideally to act as a chemically passive sink for emitted fission products. This should either effectively avoid emission from the compact or at least introduce a time delay between release from a defective coated particle and escape from the compact.

From the above-described requirements for fuel pebbles and fuel compacts, it is important to note that, due to the characteristics of pebble-bed core, i.e., flowing-down movement of pebbles and direct contact of fuel pebbles with coolant He gas and, owing to the characteristics of prismatic core, i.e., insertion of the fuel compacts into the fuel holes of the graphite block, fuel pebble surface is required for appropriate abrasiveness and corrosion and good mechanical strength, contrary to the fuel compact, for which the requirements of the mechanical integrity and corrosion rate are low. These functions are fulfilled in place by the graphite fuel blocks of the core structure. This gives the developmental works of matrix graphite materials of fuel pebble and fuel compact to have followed different directions: improved corrosion and enhanced mechanical properties for the former and thermally- and neutron-induced dimensional changes and thermal properties for the latter. [5].

Table 1 and 2 summarize the specifications of the fuel pebbles and fuel compacts, respectively, established for the Chinese HTR-10 and the Japanese HTTR. [1, 6; partly reproduced]

Table 1. Specification for Matrix graphite for HTR-10 spherical fuel elements

Inspection items	Inspection method	Specification
Reference graphite ball		
Density (g/cm^3)	Weighing and calculation	1.70
Total ash (ppm)	Burning and chemical analysis	≤300
Li content (ppm)	Burning and chemical analysis	≤0.3
Impurity B_{equ} (ppm)	Burning and chemical analysis	≤3
Thermal conductivity[a] (W/m K)	Laser pulse method	≥25
Corrosion rate[b] (mg/cm^2 h)	Corrosion furnace test/weighing	≤1.3
Erosion rate (mg/h)	Erosion device/weighing	≤6
Number of drops[c]	Free dropping device	≥50
Breaking loading crushing strength (kN)	Pressing device	≥18
CTE anisotropy ($\alpha_{\perp}/\alpha_{\parallel}$)	Thermal expansion analysis	≤1.3
Spherical fuel element		
Diameter (mm)	Visual observation	60
Thickness of fuel-free shell (mm)	X-ray project	4–6
U loading (g)	Calculation	5
U contamination	Acid leaching	
Free U fraction (U_{free}/U_{total})	Burning/acid leaching	$\leq 3 \times 10^{-4}$

[a] [1000 °C] (W/m K).
[b] [1000 °C, He + 1 vol% H_2O].
[c] Numbers of drop from 4 m high onto pebble bed before break.

Table 2. Specification for Japanese HTTR fuel compacts

Fuel compact	
Coated fuel particles packing fraction (vol.%)	30 ± 3
Impurity (ppm EBC[a])	≤ 5
Exposed uranium fraction	$\leq 1.5 \times 10^{-4}$
SiC-failure fraction	$\leq 1.5 \times 10^{-3}$
Outer diameter (mm)	26.0 ± 0.1
Inner diameter (mm)	10.0 ± 0.1
Height (mm)	39.0 ± 0.5
Matrix density (g cm^{-3})	1.70 ± 0.05
Compressive strength (N)	≥ 4900

4. Material Behavior of Matrix Graphite for Fuel Pebbles and Fuel Compacts upon Irradiation

Data have been extensively obtained from neutron irradiation experiments for fuel pebbles and fuel compacts during last 3 decades, for which the relevant as-fabricated material data are hardly traced in the literature. In this section, examples of the material behaviors are discussed on the matrix graphite material for fuel pebbles and compacts upon irradiation.

Fig. 4 (a) and (b) show the dimensional change and change in Young's modulus, respectively, for quasi-isostatically pressed matrix graphite material which is a mixture of natural and artificial graphite powders with phenolic resin binder (mixing ratio being 64:16:20 in wt%), so-called A3-3, which is used for current HTR in both fuel pebble and fuel compact fabrication. [7] Also shown are those for matrix graphite tested in the Dragon Project in Fig. 5 and 6, respectively, the material being uni-axially pressed petroleum coke graphite compacts. [8] These figures show clearly the differences in dimensional change and change in Young's modulus of two different matrix graphite materials consolidated by quasi-isostatic pressing and uni-axial pressing methods. This difference in material behavior can be, on one hand, from the material itself and, from the methods of pressing fuel pebbles or fuel compacts, on the other.

Recently, Seo et al. [9] measured Vickers hardness of the uni-axially pressed A3-3 matrix graphite material along two different directions; perpendicular and parallel to uni-axial pressing direction. The result showed that the hardness values obtained from the two directions showed an anisotropic behavior: The values obtained from the perpendicular section showed much higher micro hardness (176.6±10.5MPa in average) than from the parallel section ((125.6±14.2MPa in average). This anisotropic behavior was concluded to be related to the microstructure of the matrix graphite. This may imply that the uni-axial pressing method to make compacts influence the microstructure of the matrix and hence the material properties of the matrix graphite.

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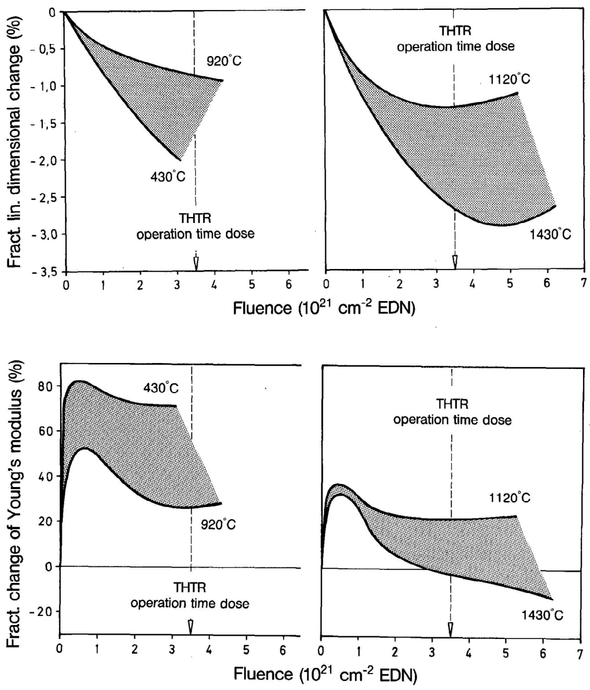
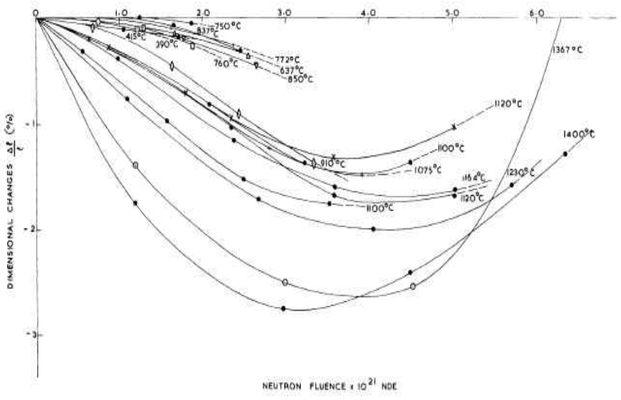
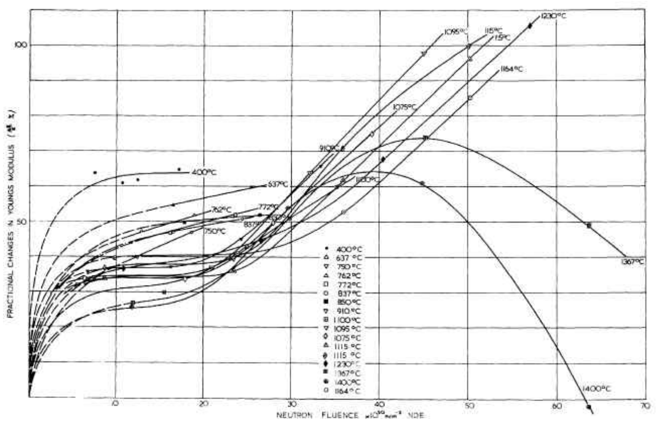


Fig. 4. Dimensional change (a) and change in Young's modulus (b) of the quasi-isostatically pressed A3-3 as a function of neutron fluence in different temperature range.



(a) Axial direction



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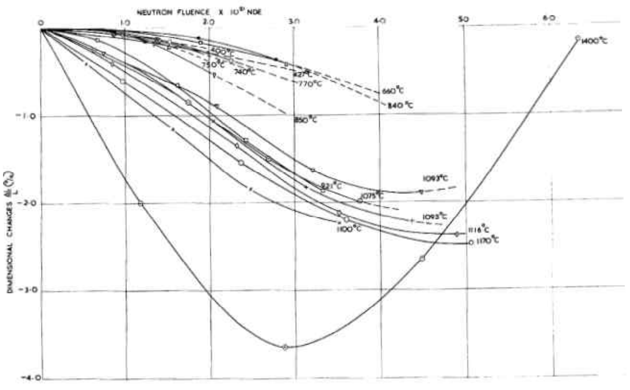
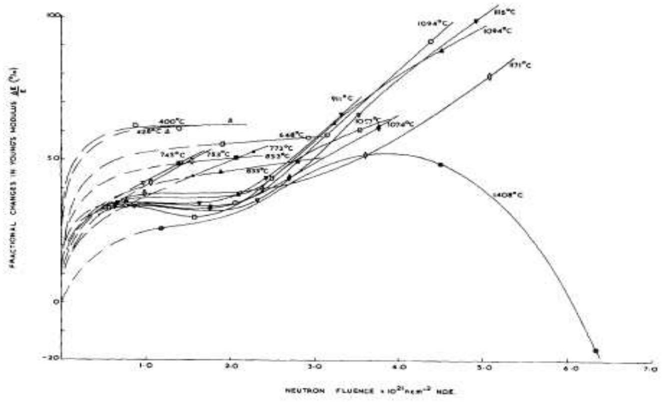


(b) Radial direction



(b) Radial direction

Fig. 5. Dimensional changes of pressed petroleum coke graphite tested in the Dragon Project. (a) axial direction and (b) radial direction in different temperatures

Fig. 6. Change in Young's modulus of pressed petroleum coke graphite tested in the Dragon Project. (a) axial direction and (b) radial direction in different temperatures