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A COMPARISON OF THE CHARACTERISTICS OF GRAPHITES IRRADIATED AT 600 and 900°C*

C. R. Kennedy,† W. P. Eatherly,† D. Minderman‡

†Oak Ridge National Laboratory, Metals and Ceramics Division,
P. O. Box X, Oak Ridge, Tennessee 37830

‡KFA, Julich, Federal Republic of Germany

Graphite for the reflector in the pebble-bed High-Temperature Reactor in the Federal Republic of Germany (FRG) requires exceptional irradiation resistance for economic viability. The candidate graphites are being irradiated in the High Flux Isotope Reactor (HFIR) at Oak Ridge to assess their irradiation life expectancy. This assessment is based upon dimensional stability, elastic constants from sonic testing, electrical resistivity from eddy-current testing, coefficient of thermal expansion, and brittle-ring fracture testing. To date 13 FRG grades have been irradiated and compared with four graphites made in the United States and one from the United Kingdom. The irradiation temperatures were 620, 715, and 900°C.

The dimensional changes of all of the graphites reflected a reasonably isotropic structure with only mild preferred orientations. Most grades demonstrated normal linear shrinkage with initial fluence and subsequent expansion as fluence increased (Fig. 1). The definition of lifetime expectancy is based upon the fluence required to cause a volume expansion to return the density to the unirradiated value. This has been demonstrated to be conservative with regard to the eventual loss of mechanical properties to values lower than unirradiated material. A comparison of the lifetime expectancy of a number of the grades is given in Fig. 2 showing the general trend of reduced life with increasing temperature. The more highly

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INCREASING IRRADIATION TEMPERATURE REDUCES LIFE EXPECTANCY

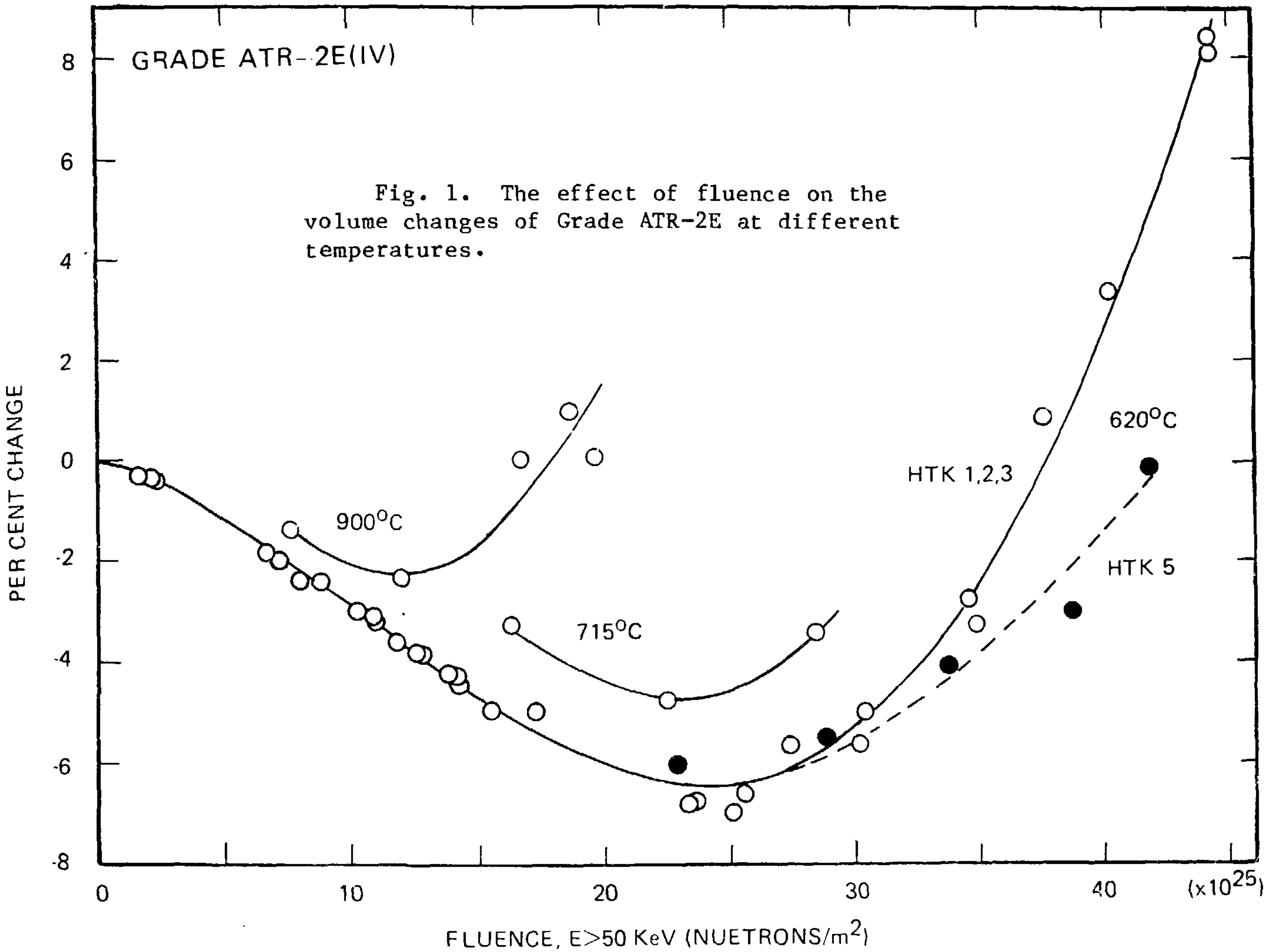

Fig. 1. The effect of fluence on the volume changes of Grade ATR-2E at different temperatures.

crystalline graphites (reduced x-ray line broadening) have a longer life expectancy. The FRG pitch coke graphites have the lowest crystallinity with the shortest life expectancy and GRAPHNOL N3M has the highest crystallinity with the highest life expectancy. This should be expected as the more crystalline materials have lower crystallite growth rates. Therefore, the internal shearing rates by differential growth will be reduced and the fluence required for structural degradation will be extended.

The dimensional stability of these isotropic graphites depends almost totally upon the densification caused by irradiation. The densification is a result of the differential growth to close the existing defect structure in the polycrystalline graphite. It can be postulated that this defect structure consists of (1) voids from poor packing and shrinkage of the binder and impregnant in processing that are external to particulate filler coke, and (2) a defect structure within the filler coke as a result of microcracking within the particles due to the anisotropic a- and c-axis crystallographic coefficients of thermal expansion during cool down from the graphitization temperature. Therefore, the total densification under irradiation can be considered to be a function of the volume created by the anisotropic coefficients of thermal expansion and the overall porosity:

$$(\Delta V/V_0)_I = f\ (\Delta V/V_0)_{CTE}\ ,\ (\Delta V/V_0)_D\ ,$$

where

$(\Delta V/V_0)_I$ = maximum densification under irradiation

$(\Delta V/V_0)_{CTE}$ = $2000\ (27 \times 10^{-6} - CTE_v)$, the void volume created by cooling down from 2000°C

$(\Delta V/V_0)_D$ = (2.05 — initial bulk density)/2.05 = the void volume within the structure accessible to helium, and 2.05 is the apparent helium density.

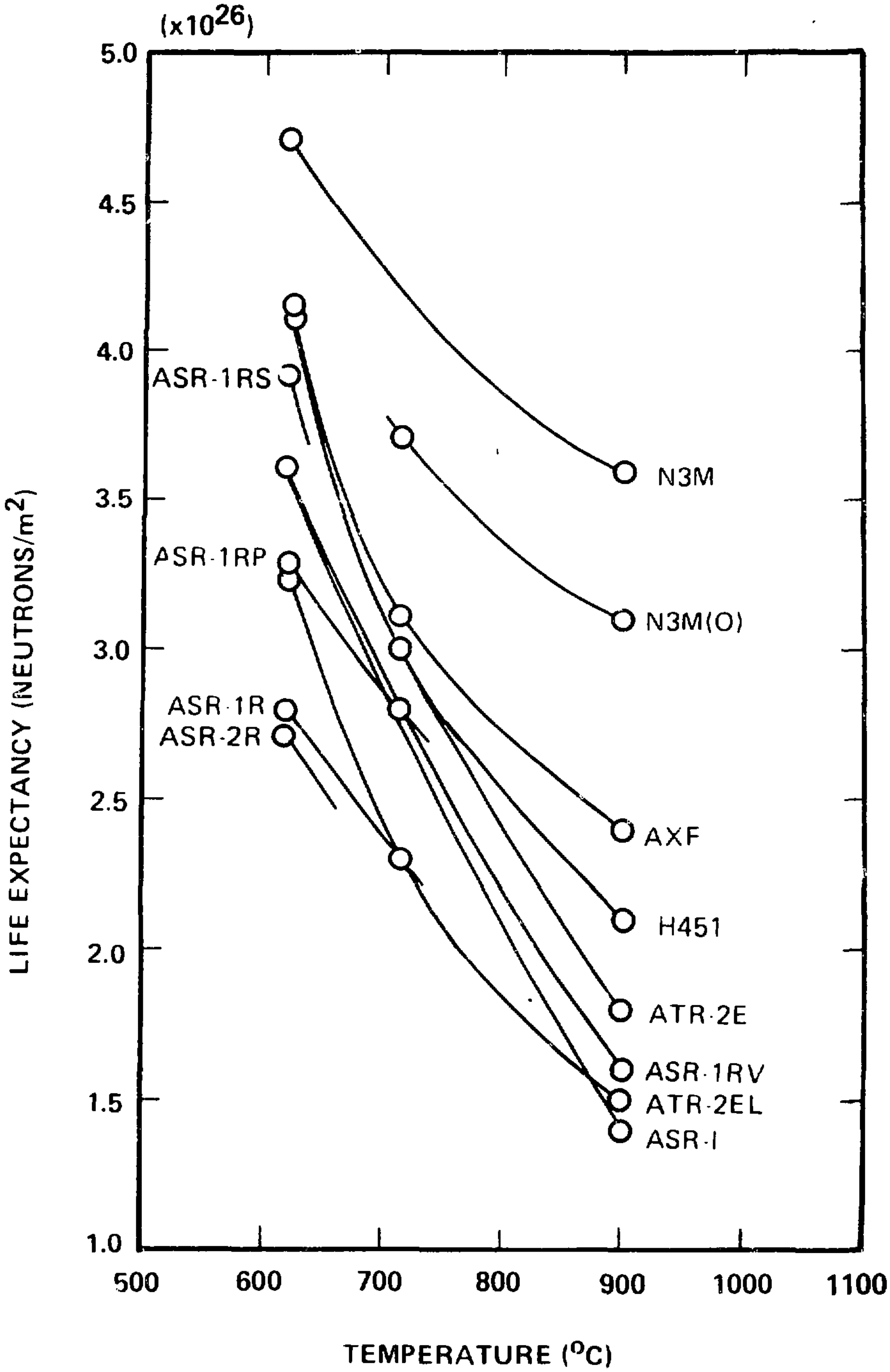

Fig. 2. The effect of irradiation temperature on the life expectancy of graphite under irradiation.

Now that we have irradiated a reasonable number of grades with a range of $(\Delta V/V_0)_I$, it would be relevant to examine the data to determine the relative significance of $(\Delta V/V_0)_{CTE}$ to $(\Delta V/V_0)_D$. This should also allow the prediction of the maximum densification of future materials. As a first approximation and for simplicity, we assume a linearized function for a sensitivity calculation in the form

$$(\Delta V/V_0)_I = A + B \ (\Delta V/V_0)_{CTE} + C \ (\Delta V/V_0)_D \ .$$

A multiple linear regression analysis yields the coefficient values A = −0.0825, B = 2.85, and C = 0.85 at 620°C irradiation temperature. The comparison of measured and computed values are given in Table I.

Table I. A comparison of actual to calculated maximum densification (irradiation temperature = 620°C)

Grade	Actual $\frac{\Delta V}{V_0}$, %	Calculated $\frac{\Delta V}{V_0}$, %
V356	9.6	9.2
V483	9.1	8.1
ATR-2R	7.0	7.1
ATR-2E	7.1	7.1
ATR-2EL	6.5	8.0
ASR-1R	5.3	5.9
ASR-1RP	6.7	7.4
ASR-1RS	7.1	7.8
ASR-2R	8.5	8.4
UKAEA-11	5.3	5.2
AXF	1.6	1.4
H451	9.9	9.3

We can conclude that at 620°C the thermal expansion voids are three times (B = 2.85) more effective than their intrinsic volume in controlling the irradiation induced densification, whereas the helium-accessible voids are effective on a one-to-one ratio (C = 0.85). We also note that the quantity $(\Delta V/V_0)_I$ changes by 3 to 4 V/o as the irradiation temperature changes from 620 to 900°C as shown in Fig. 3. This suggests the coefficient B should be ~10 rather than the 2.85 we calculate at 620°C. Quite clearly the linearized sensitivity equation we

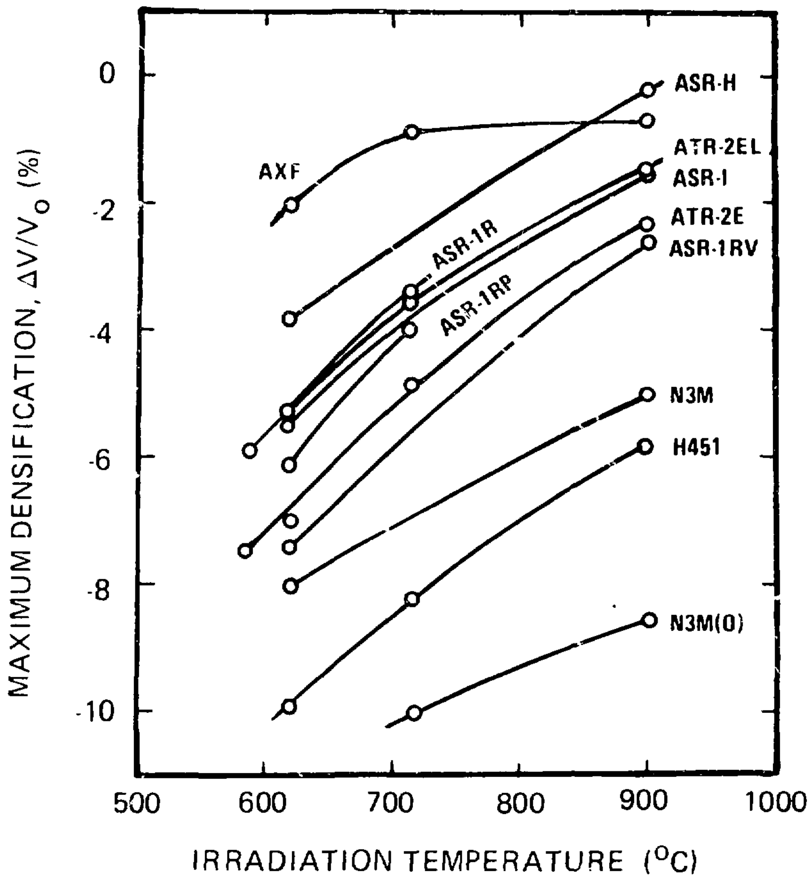

Fig. 3. The effect of irradiation temperature on the maximum densification of graphite.

have fitted successfully at 620°C must at the very least have temperature-dependent coefficients. The major cause of the lifetime reduction is the loss of void volume by thermal expansion to accommodate the differential anisotropic growth of irradiation. The thermal expansion voids are between 3 and 10 times as effective in reducing the potential densification by irradiation.

The electrical resistivity from eddy-current measurements demonstrated a rapid saturation in resistivity by irradiation followed by a slight reduction with increasing density. After maximum densification is reached, the resistivity begins to increase as the structure is degraded. The effect of temperature appears to be nominal until the structure degrades.

The modulus of elasticity as calculated from longitudinal and shear velocities appears to be in conflict with data in the literature for low fluences. The modulus of elasticity appears to increase more rapidly with increasing irradiation temperature, reaches a lower maximum value, and begins to decrease more rapidly with structural degradation.

In summary these experiments have been successful in yielding estimates of lifetime as a function of temperature. It was shown that the more crystalline grade GRAPHNOL N3M yielded the highest life expectancy of all grades. The overall dimensional stability can be reasonably estimated from the unirradiated density and the CTE. The implication of these results is that the reduction in life with increasing temperature is a result of the loss of accommodating volume by the anisotropic a- and c-axis crystallographic coefficients of thermal expansion.