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ABSTRACT

Experimental data on the mechanical strength of nuclear graphite subjected to fast neutron irradiation are reviewed. At fluences below the "turnaround" point, the mean tensile, flexural, or compressive strength, S, increases over the unirradiated value, S_o, in a manner related to the irradiation-induced change in Young's modulus, E:

$$\frac{S}{S_o} = \left(\frac{E}{E_o}\right)^k$$

The exponent, k, takes a value between 0.5 and 1, depending on the graphite grade, the irradiation temperature, and the method used for determining E. A value of 0.5 for k would be expected from the Griffith-Irwin theory of fracture if neither the critical flaw size nor the effective surface energy of a crack is altered by irradiation; a value of 1 would be expected if the strain at failure remains constant. At higher fluences, when expansion starts, strength values decrease. The effect of irradiation on the statistical spread of the strength measurements depends on the graphite grade and the neutron fluence. At fluences below the "turnaround" point, the coefficient of variation of strength determinations for standard reactor grades is little changed, but at higher fluences, and for fine-grained, high strength materials, the coefficient of variation may increase. Although specimens for different locations in a graphite log, or from different logs of the same grade, vary in strength, under the same conditions of irradiation the fractional increase in strength of these specimens will be similar. After irradiation, graphite appears to have a slightly greater resistance to cyclic fatigue, as measured by the homologous stress limits for survival to a specified number of cycles.

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1. INTRODUCTION

Graphite components in nuclear reactors are subject to both primary stresses caused by dead loads and differential gas pressures, and secondary stresses arising from gradients in temperature and neutron fluence. Safe design of these components requires full understanding of the mechanical strength of graphite under the appropriate conditions. In this paper experimental data from the U.S. and Europe on the mechanical strength of irradiated nuclear graphite are briefly reviewed. The topics include the change in mean strength as a function of neutron fluence and irradiation temperature; the relationship between irradiation-induced changes in strength and Young's modulus; changes in the statistical scatter of the strength measurements; changes in the strength of specimens from different log locations or different logs of the same grade; and the effect of irradiation on cyclic fatigue strength.

2. IRRADIATION-INDUCED CHANGES IN MEAN STRENGTH

At neutron fluences below the point where volume shrinkage ceases (the "turnaround" point), neutron irradiation increases the mean strength of graphite, whether measured in tension, compression, or bending. A typical plot showing the change in the compressive strength of a reactor grade graphite, taken from the work of Platanov, et al,[1] is shown in Figure 1. At the low temperatures and fluences used in this work the strength undergoes a rapid increase to a plateau, whose level decreases with increasing irradiation temperature. At this point, the strength is from 20 to 150% higher than the preirradiation value, depending on the irradiation temperature. At higher neutron fluences the strength shows a second, more gradual increase, accompanied by a similar increase in Young's modulus. Figure 2, taken from UKAEA data on near isotropic graphites irradiated in the Dounreay Fast Reactor,[2] shows the strength changes on specimens carried to a high fluence. Finally, the "turnaround" point is reached and the specimen expands, generating new internal porosity. At this point, strength measurements start to show a marked decrease, as is apparent in Figure 2.

The majority of the strength data reported in the literature show the trends summarized in the previous paragraph. Tensile tests conducted at General Atomic[3] on specimens of H-451, H-327, and TS-1240 graphites irradiated at temperatures between 600°C and 1350°C to maximum fluences of 11 x 10^{25} n/m^2 (equivalent fission fluence for graphite damage) showed consistent increases in strength of up to 100%. Tensile tests on the pitch-coke graphite AS2-500 irradiated to a peak fluence of about 12 x 10^{25} n/m^2 (equivalent fission fluence) between 425°C and 1005°C, reported by KFA workers,[4] showed a steady upward trend in strength as the fluence increased, with the mean strengths tending to be higher for lower irradiation temperatures. Diametral compression tests (Carneiro tests) on discs of the same material irradiated at 500°C to much lower fluences (up to 1 x 10^{25} n/m^2) also showed the expected rise in strength.[5] Four-point bend tests on a fine-grained, high-strength graphite (Poco grade AXF-5Q) irradiated at 400°C to a peak fluence of 3 x 10^{25} n/m^2 (E > 1 MeV) displayed similar trends,[6] as did bend tests on gilsocarbon-based graphites irradiated in the Dragon reactor between 850° and 1250°C to fluences between 0.5 and 5 x 10^{25} n/m^2 (equivalent fission fluence).[7] Somewhat different trends were noted during "brittle-ring" tests (diametral compression of a hollow ring) on fine-grained, high-strength graphites irradiated at ORNL under the Molten Salt Breeder Reactor development program.[8,9] Specimens of grades AXF, AXF-5QBG-3, H-395, and P-03 graphites were irradiated in the HFIR reactor at 715°C to various fluences up to 40 x 10^{25} n/m^2 (equivalent fission fluence). The "brittle-ring" strengths are shown as a function of neutron fluence in Fig. 3; after an initial increase, the strengths decline with increasing fluence. In comparison with the UKAEA data (Figure 2), the decline in strength for the fine-grained graphites appears to start at a lower fluence.

One set of data reported in 1970 appeared to show a serious strength reduction at quite low neutron fluences. Lungagnani and Krefeld[10] reported bend strength data on the gilsocarbon graphite irradiated in the Petten HFR at 600°C and at 1150°C. While the 600°C data showed the expected increase in mean strength with increasing neutron fluence, at 1150°C the mean strength first increased and then fell by more than 50% for a neutron fluence of only 1.2 x 10^{25} n/m^2 (equivalent fission fluence). However, it is now believed that the 1150°C results were caused by water vapor in the capsule oxidizing the graphite, and this set of data should be disregarded.

3. RELATIONSHIP BETWEEN CHANGES IN STRENGTH AND YOUNG'S MODULUS

Graphite is a brittle solid which fails by the propagation of cracks originating at pre-existing flaws. According to the Griffith-Irwin model of brittle fracture, the tensile strength, S, is related to Young's modulus, E, the maximum flaw size, a, and the effective work of fracture per unit crack area, γ, by the expression:

$$S = A\sqrt{\frac{E\gamma}{a}} \qquad (1)$$

where A is dimensionless constant related to the flaw geometry. A process which changes the value of Young's modulus without altering the value of (γ/a) should therefore change the strength according to the relationship:

$$\frac{S}{S_o} = \sqrt{\frac{E}{E_o}} \qquad (2)$$

This relationship also implies that the strain energy at failure remains unchanged. Losty and Orchard[11] observed that neutron irradiation at low temperatures and fluences increased both Young's modulus and strength in the ratio predicted by Eq. (2); similarly, heat treatment of ungraphitized carbons reduced Young's modulus and the strength while maintaining a constant strain energy at failure. The conclusion of this work was that low-fluence neutron irradiation or heat treatment changes Young's modulus of the crystallites but does not affect the pores or microcracks between them.

A constant strain energy at fracture was also observed with Poco AXF-5Q graphite irradiated at 400°C,[6] using a resonant bar technique to measure Young's modulus and four-point bend tests to measure strength. The UKAEA data plotted in Figure 2 also agree well with Eq. (2) up to the point where the strength starts to decrease. On the other hand, an increase in compressive strength considerably greater than predicted by Eq. (2) was observed by Platanov, et al.[1] in reactor graphite irradiated at 90° - 150°C, while at 400° - 500°C, the increase in strength of reactor graphite was about as predicted. In the same experiment, the fractional strength increase of ungraphitized carbon was similar to that of reactor graphite, whereas the fractional increase in Young's modulus of the carbon was only about half the increase for graphite. Taylor, et al.[12] found that the Young's modulus of three types of isotropic graphite increased by about 100% when irradiated at 150°C to about 10^{25} n/m^2; the tensile strength increase in one of these graphites was about 50%, about as expected from Eq. (2), but the strength increase in the other two graphites was nearer 100%. Increases in compressive strength were about 200% in all three graphites. It is evident that at low irradiation temperature there are many deviations from the constant strain energy rule, and that the type of graphite exerts a strong influence. It is worth noting that at these low irradiation temperatures, crystallite c-axis expansion tends to close up microcracks at an early stage, which could result in a reduction of the critical flaw size [a in Eq. (1)].

At higher irradiation temperatures, too, irradiation-induced strength increases often exceed the levels predicted by Eq. (2). Everett and Ridealgh[7] measured the tensile and bend stress-strain properties and the dynamic Young's modulus of gilsocarbon based graphites irradiated in the Dragon reactor at 900 -1250 C to a maximum fluence of 5 x 10^{25} n/m^2 (equivalent fission fluence). They pointed out that, because of the non-linear stress-strain curve, it is important to specify how Young's modulus is defined. When dynamic measurements were used, their strength (S) and Young's modulus (E) data fitted the relationship:

$$\frac{S}{S_o} = \frac{E}{E_o} \tag{3}$$

Even so, there was some reduction in total strain to failure. They suggested that the elastic component of the strain to failure remains constant during irradiation. During tensile tests conducted at General Atomic Company on specimens of H-451, H-327, and TS-1240 graphites,[8] Young's modulus was determined from extensometer measurements. Irradiation temperatures ranged between 590°C and 1350°C, and fluences ranged up to 11 x 10^{25} n/m^2 (equivalent fission fluence). The strength data fell between the levels predicted by Eq. (2) and Eq. (3). The data could conveniently be represented by the expression:

$$\frac{S}{S_o} = \left(\frac{E}{E_o}\right)^k \tag{4}$$

where k is a constant between 0.5 and 1 which depends on the graphite grade. For H-451 graphite, on which most data were obtained, k had the value 0.67.

One may conclude that the constant strain energy relationship (Eq. 2) gives a generally conservative guide to the strength of irradiated graphite up to the point of "turnaround." Where sufficient data are available for a particular graphite grade and irradiation condition, Eq. (4), with an empirically determined value for k, will be more accurate.

4. STATISTICAL SCATTER IN THE STRENGTH MEASUREMENTS

The statistical scatter in strength determinations on a brittle material is just as important to design as the mean strength. Matthews' bend tests on groups of 26-29 specimens of irradiated Poco AXF-5Q graphite[6] showed a definite increase in scatter, with the Weibull modulus decreasing from 16 for unirradiated material to about 10 for irradiated specimens (corresponding to an increase in coefficient of variation from about 7% to 11%). Tensile data on irradiated AS2-500 graphite reported by Schiffers, et al.[4] showed a wide scatter band, but the data were not analyzed statistically. Other workers have found that, while the standard deviation increases, the coefficient of variation remains unchanged. The coefficient of variation based on sets of 8-12 tensile strength determinations on H-451 graphite[3] irradiated between 590°C and 1350°C is shown as a function of neutron fluence in Figure 4. There is no indication of an increase. Brocklehurst[2] also reported that the coefficient of variation of bend strength determinations on IM1-24 graphite remained unchanged at 8% following irradiation at 900°C to 4.5×10^{25} n/m^2 (equivalent fission fluence). Carneiro tests on groups of 34 irradiated specimens of pitch coke graphite[5] showed no increase in the range of the determinations, which implies a slight reduction in the coefficient of variation. Thus, the bulk of the evidence points towards the coefficient of variation of strength determinations remaining unchanged with irradiation, at least up to the "turnaround" point. The only apparent exception is the fine-grained, high-strength Poco graphite AXF-5Q.[6]

5. WITHIN-LOG AND BETWEEN-LOG VARIATIONS

For given irradiation conditions, the fractional increase in strength is likely to be different for different types of graphite, since the irradiation-induced increase in Young's modulus is structure-dependent. In addition, Taylor, et al.[12] found that one of three isotropic graphite grades irradiated at 150°C experienced greater strengthening than the other two, even through the changes in Young's modulus were similar. In contrast, irradiation-induced fractional increases in the strength of graphite specimens from different locations in the same log, or from different logs of the same grade, appear to be similar, despite the fact that the mean strengths may vary by more than a factor of 2. Figure 5 shows the fractional increase in tensile strength of H-451 graphite irradiated at about 900°C. The data include specimens from three different production lots and two log locations (midlength-center and midlength-edge), as well as both axial and radial orientations. There is no obvious difference in the fractional strength increases, even though the unirradiated tensile strengths of the different specimen groups ranged from 8.5 to 18.6 MPa.

6. EFFECT OF IRRADIATION ON CYCLIC FATIGUE STRENGTH

Graphite is subject to cyclic fatigue in a way similar to many metals. Brocklehurst[2] showed a plot of the homogous stress (peak tensile stress during the fatigue cycle divided by the mean strength) versus the number of cycles to failure for IM1-24 graphite tested in a variety of loading modes, which included some bend specimens irradiated at 900°C to 4.5×10^{25} n/m^2 (equivalent fission fluence). The data points for irradiated graphite fell in the same scatter band as the unirradiated data. In a more systematic study at General Atomic Company,[13] three groups of 42 tensile specimens of H-451 graphite were irradiated at 900°-990°C to 3.8, 7.1, and 10.6 x 10^{25} n/m^2 (equivalent fission fluence). After irradiation, 10 specimens from each group were tensile tested to establish the mean tensile strength and the remainder were fatigue tested with reversed stress (tension-compression) cycling. The homologous stress limits for survival to 10^5 cycles were substantially higher than for unirradiated specimens, with the exception of one group of radial specimens, whose homologous stress limits were similar to those of their unirradiated companions. Figure 6 shows the stress limits for survival to 10^5 cycles plotted as a function of neutron fluence. The stress limits in Figure 6 were normalized by dividing by the mean unirradiated tensile strength, so that the rise in fatigue strength is partially due to the irradiation-induced increase in tensile strength. In addition to the curve for 50% specimen survival, Figure 6 also includes a curve for 99% specimen survival (at 95% confidence). The letter curve shows a slight downtime at the highest fluence which is attributable to an increase in data scatter. It is evident from Figure 6 that the irradiation induced improvement in tensile strength also applies to the cyclic fatigue strength.

7. CONCLUSIONS

(1) Up to the "turnaround" point, the tensile, compressive, and flexural strength of graphite increases with neutron fluence in a manner which parallels the increase in Young's modulus.

(2) Beyond the "turnaround" point, the strength declines.

(3) The increase in strength, S, is related to the increase in Young's modulus, E, by the expression:

$$\frac{S}{S_o} = \left(\frac{E}{E_o}\right)^k$$

where k is a constant, between 0.5 and 1, whose value depends on the type of graphite and the irradiation conditions.

(4) The irradiation-induced strength increases are qualitatively consistant with the Griffith-Irwin model for the fracture of brittle materials. At intermediate temperatures and fluences, Young's modulus of the crystallites increases while the flaw size remains unchanged, resulting in a k-value of 0.5. In the absence of sufficient strength data, assuming a value of 0.5 will generally result in conservative strength predictions.

(5) The coefficient of variation of strength determinations may be assumed to remain constant with irradiation, with the possible exception of fine-grained, high strength graphites where the statistical scatter may increase.

(6) Under the same irradiation conditions, the fractional increase in strength of specimens from different log locations, or from different logs of the same graphite grade, will be similar.

(7) The irradiation-induced increase in tensile strength is retained or improved under cyclic fatigue conditions.

8. REFERENCES

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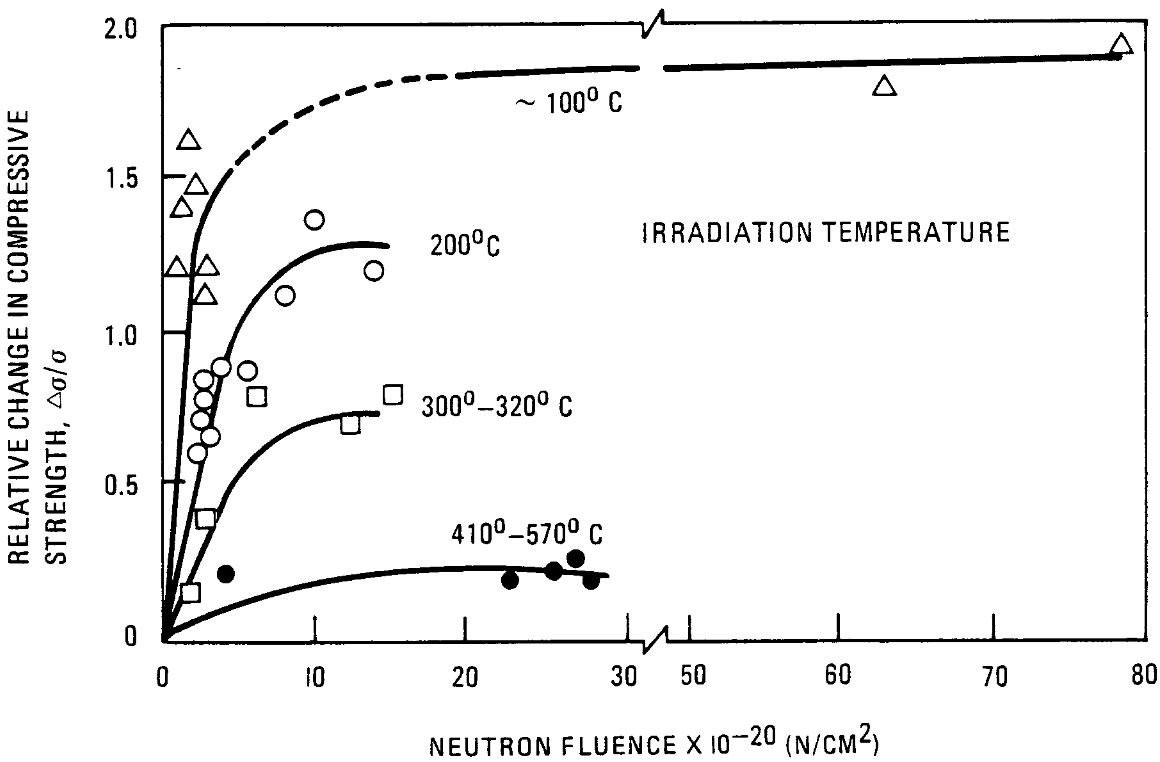

Fig. 1. Relative change in compressive strength of reactor graphite versus integral flux (from Ref. 1)

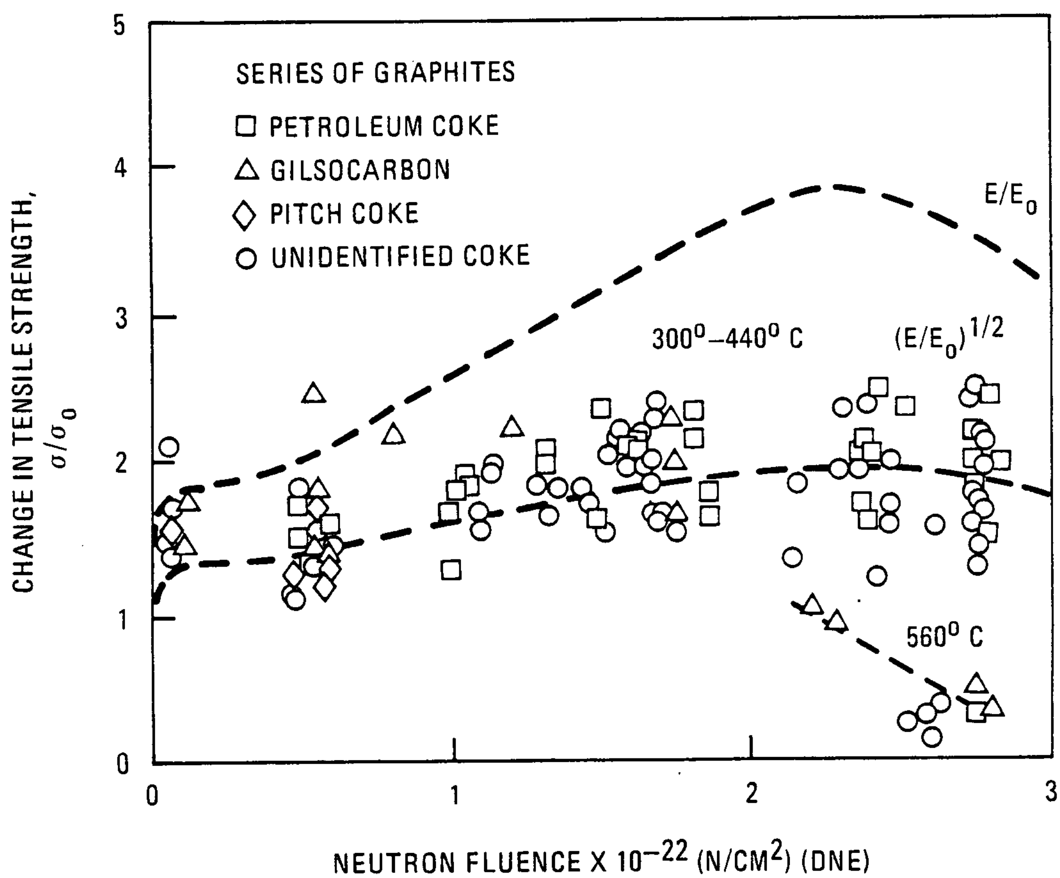

Fig. 2. Tensile strength changes with fast neutron dose for different near-isotropic graphites irradiated in DFR at 300° to 440°C and at 560°C (from Ref. 2)

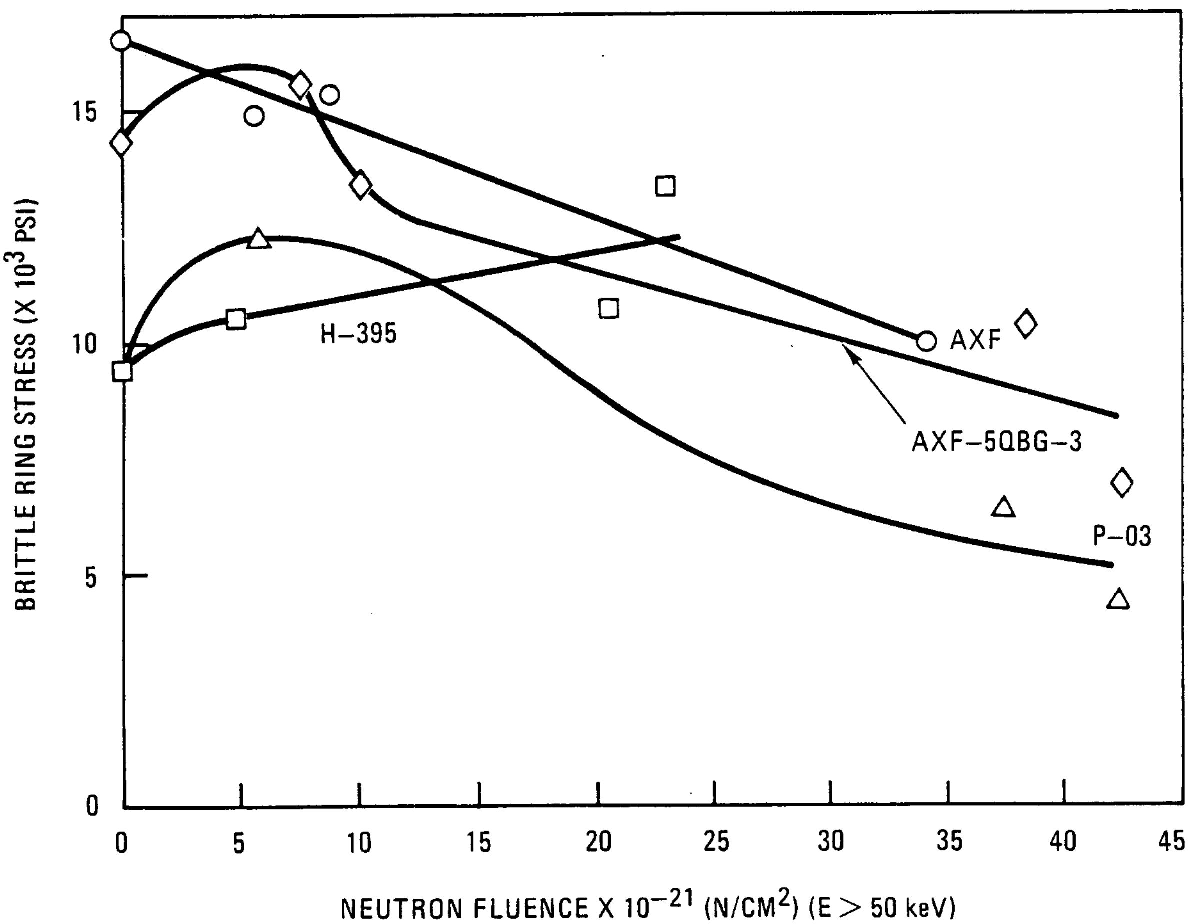

Fig. 3. Brittle ring stresses versus fluence accumulated at 715°C for graphite grades AXF, AXF-5QBG-3, H-395, and P-03 (from Ref. 8)

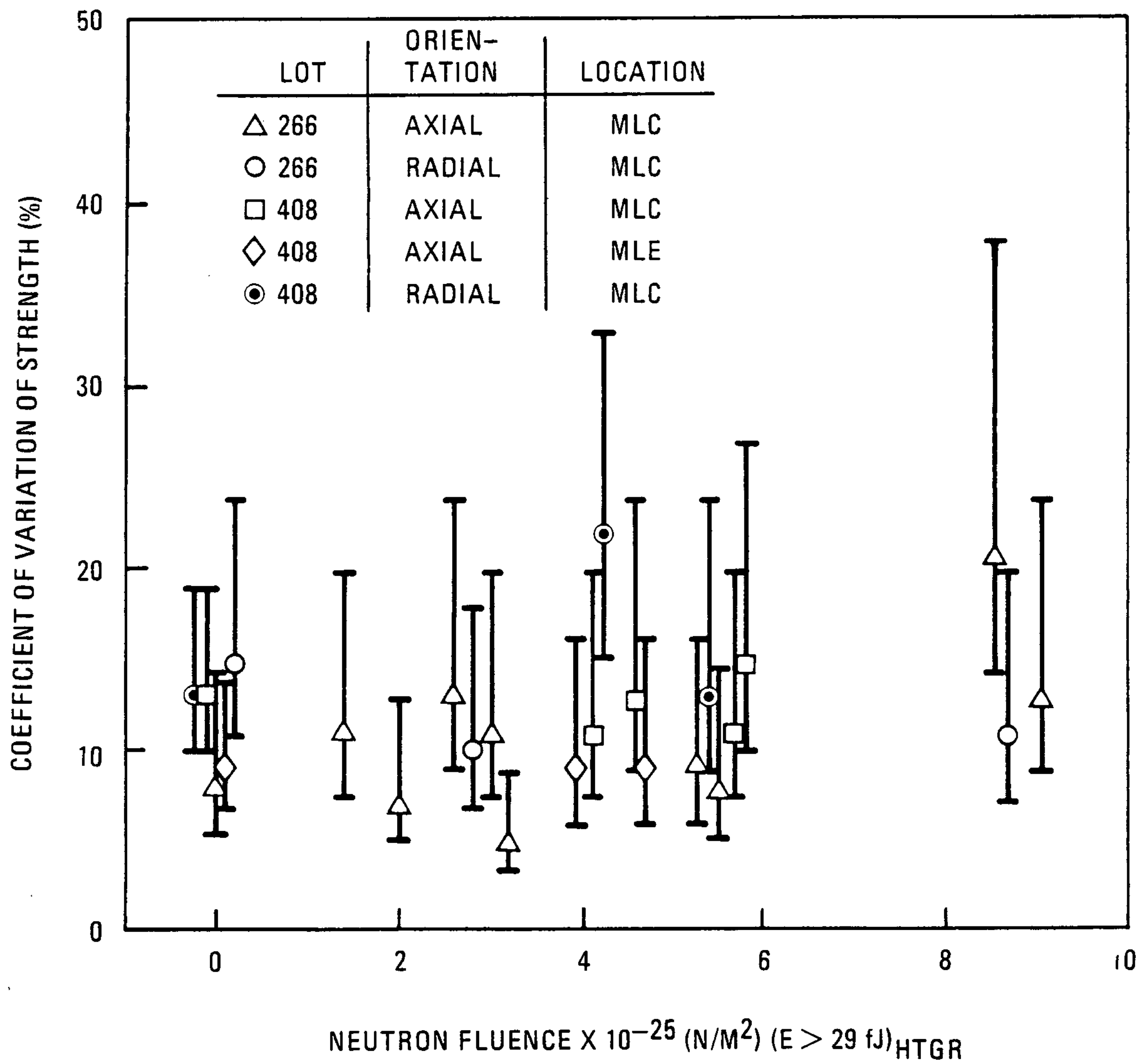

Fig. 4. Coefficient of variation of the tensile strength of H-451 graphite as a function of fast neutron fluence (from Ref. 3). Error bars denote 95% confidence interval.

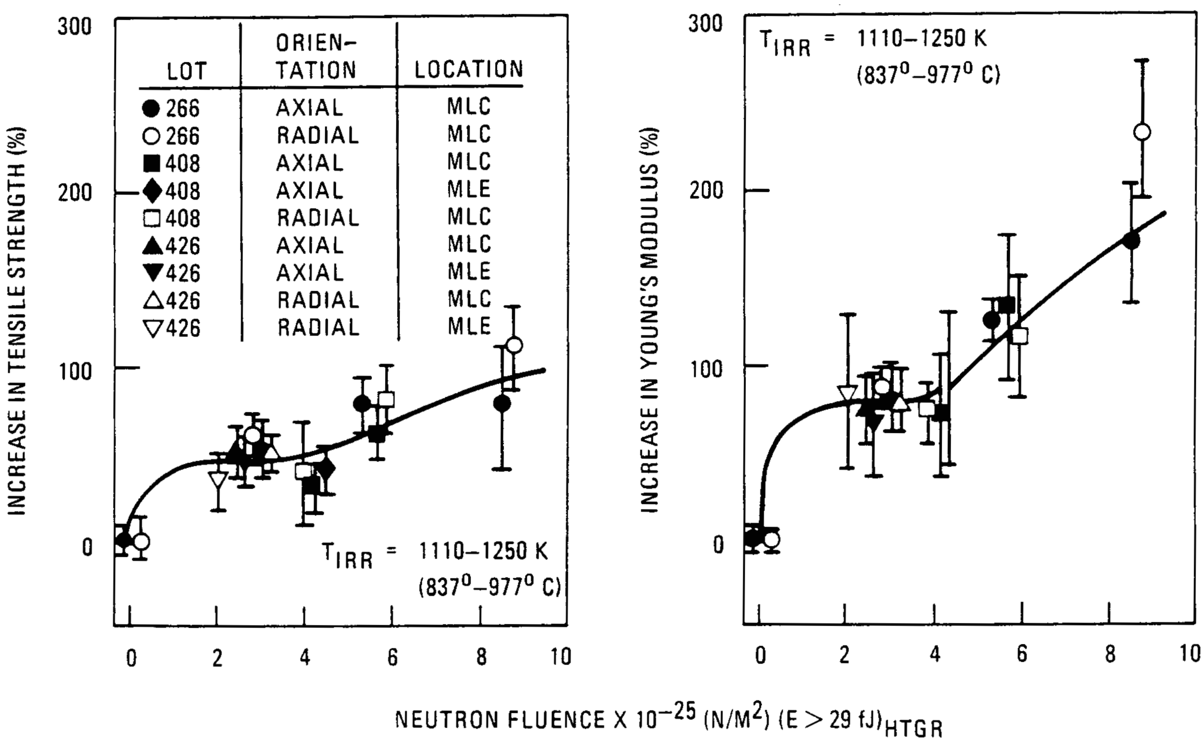

Fig. 5. Changes in tensile strength and Young's modulus of H-451 graphite as a function of fast neutron fluence (from Ref. 3). Error bars denote ±1 standard deviation.

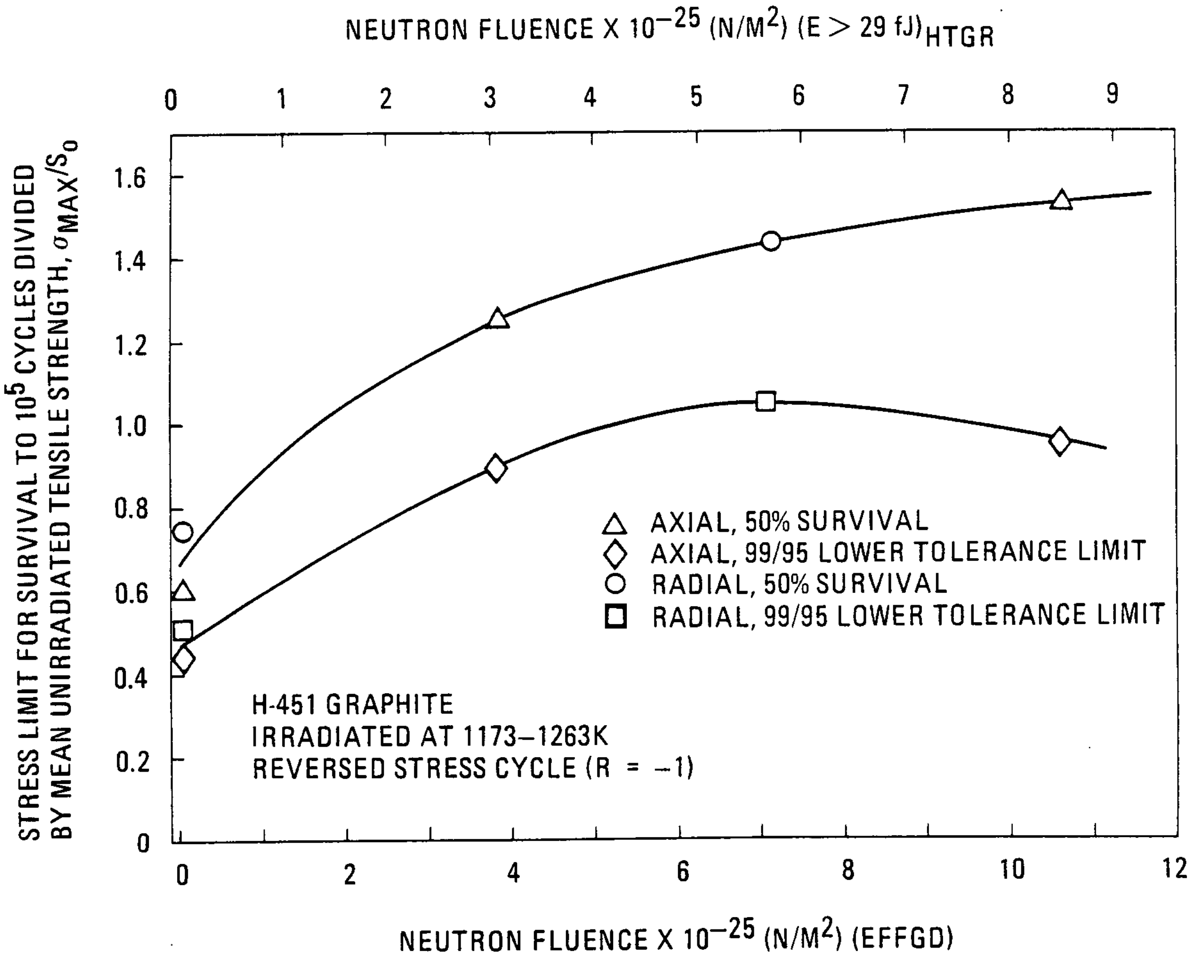

Fig. 6. Fatigue stress limits for survival of H-451 graphite to 10^5 cycles, normalized to the unirradiated tensile strength, versus the fast neutron fluence at an irradiation temperature of 900° to 990°C (from Ref. 13)