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EFFECTS OF FAST-NEUTRON DAMAGE FROM 0 TO 42 × 10^{21} NEUTRONS/CM^2 ON THE PHYSICAL PROPERTIES OF NEAR-ISOTROPIC GRADES OF GRAPHITE*

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The characterization of property changes in various grades of near-isotropic, "binderless" grades of graphite as functions of fluence accumulated at 715°C from 0 to 42 × 10^{21} neutrons/cm^2 (E > 50 keV) was made. Generally, the average coefficients of thermal expansion (CTE) from 20 to 600°C and the room-temperature values for strengths, fracture strains, Young's moduli, shear moduli, and calculated figures of merit (FOM) for resistance to thermal shock all ultimately decreased with fluence.

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*Research sponsored by the Energy Research and Development Administration under contract with the Union Carbide Corporation and the Air Force Materials Laboratory/MXS of the Wright-Patterson AFB.

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Past and Current Studies

The characterization of property changes in various grades of graphite as functions of fluence accumulated at 715°C has been a continuing part of the graphite studies for the Molten Salt Reactor Program (MSRP). In these studies, almost all of the specimens, nominally 0.126-in.-ID by 0.400-in.-OD by 0.500-in. long, have been irradiated in target pin locations in the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory.

The unirradiated (control) data of the specimens were determined. After they had been irradiated, their dimensional and volume changes were determined as functions of the fluence that they had accumulated. Other postirradiation data such as BET surface area, helium densities, open-pore porosities, and layer heights were measured for some grades of graphite. Recently, we have been able to extend this substantial quantity of experimental work through funding from the Air Force Materials Laboratory. These additional data acquired on the MSRP specimens as functions of accumulated fluence are thermal expansivity from room temperature to 625°C and mechanical properties measured at room temperature. The mechanical properties included brittle-ring strengths, fracture strains, Young's moduli, shear moduli, and calculated thermal shock resistivities.

The purpose of the studies for the Air Force Materials Laboratory was to determine if the reduction of the coefficient of thermal expansion (CTE) of a high-strength, near-isotropic grade of graphite by neutron damage would improve the relative thermal shock resistance of graphite.

Materials

For this study, graphite grade AXF was chosen because of its near isotropy and high-strength properties. We included in this study other near-isotropic grades; principally, grades AXF-5QBG-3 (grade AXF-5QBG fired at 3000°C for 1 hr), H-395, and P-03, to acquire additional information associated with the aerospace applications and nuclear reactors such as the Molten-Salt Breeder Reactors (MSBRs) and High-Temperature Gas-Cooled Reactors (HTGRs). All of these are of "binderless" grades of graphite and all tend to have relatively uniform distribution of small pores except for grade P-03. The porosity for grade P-03 appears to be larger and concen-

Tests

For the AFML/MXS work, we used the MS specimens that were nominally 0.126 in. ID 0.400 in. OD × 0.500 in. long that had bee irradiated in the High Flux Isotope Reacto (HFIR) at 715°C to various fluences up to high as 42 × 10^{21} neutrons/cm^2 (E > 50 ke The initial (Young's) and shear moduli we determined on these existing specimens by sonic techniques. The thermal expansion p ties of the specimens in the directions of their axes of rotation were determined fr room temperature to 625°C in a quartz dila eter. These measurements concluded the ne sary nondestructive testing of the specime The flexural strength tests were destructi and were obtained using the brittle-ring t nique.

Property Changes as Functions of Fluence

The maximum accumulated fluence for g AXF is 34 × 10^{21} neutrons/cm^2; those for t other grades are 23 to 42 × 10^{21} neutrons/ These fluences are 1.5 to 2.5 times that c rently attained by other investigators fo these grades of graphite. The volume cha of these grades of graphite are plotted a tions of fluence and have been reported previously.[1,2]

To avoid annealing out any of the ne damage acquired by the specimens at the i tion temperature of 715°C, the maximum te ture permitted in the quartz dilatometer the thermal expansivity determinations fo specimens was 625°C. The average coeffici of expansion (CTE) values for 20 to 600°C plotted versus fluence show that the resul the neutron damage produced similar change these grades of graphite. All show an in and then a decrease in magnitude that appe to saturate between approximately 3 to 4 × °C^{-1} for all fluences greater than 20 × 1 neutrons/cm^2 and continues this way at le through 40 × 10^{21} neutrons/cm^2. In studi for fluences through 8 × 10^{21} neutrons/cm acquired at 400 to 700°C, Pitner[3] also fo the initial CTE increase with fluence in g ite grade AXF-8Q1, a slightly more graphi form of grade AXF. In our study, the cha in the average CTE values for grades AXF, 5QBG-3, H-395, and P-03 versus fluence sh that the maximum CTE increases ranged from from 10 to ~50%. Beyond 20 × 10^{21} neutro the CTE values for all grades except gra

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Tests

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To avoid annealing out any of the neutron damage acquired by the specimens at the irradiation temperature of 715°C, the maximum temperature permitted in the quartz dilatometer during the thermal expansivity determinations for the specimens was 625°C. The average coefficient of expansion (CTE) values for 20 to 600°C plotted versus fluence show that the results of the neutron damage produced similar changes in these grades of graphite. All show an increase and then a decrease in magnitude that appears to saturate between approximately 3 to 4 × 10^{-6} $°C^{-1}$ for all fluences greater than 20 × 10^{21} neutrons/cm^2 and continues this way at least through 40 × 10^{21} neutrons/cm^2. In studies for fluences through 8 × 10^{21} neutrons/cm^2, acquired at 400 to 700°C, Pitner[3] also found the initial CTE increase with fluence in graphite grade AXF-8Q1, a slightly more graphitic form of grade AXF. In our study, the changes in the average CTE values for grades AXF, AXF-5QBG-3, H-395, and P-03 versus fluence show that the maximum CTE increases ranged nominally from 10 to ∿50%. Beyond 20 × 10^{21} neutrons/cm^2 the CTE values for all grades except grade P-03

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decreased to within a range of 40 to 60% of their initial values. Grade P-03 returned to only slightly less than its original value with the suggestion that it is slowly decreasing with fluence.

The greatest reduction in CTE value was that of approximately 60% for grade AXF. For essentially the same grade of graphite, AXF-Q1, Pitner reported a CTE decrease approaching 80% for a fluence of 16 × 10^{21} neutrons/cm^2 accumulated at 1100 to 1300°C (ref. 4). It will be shown later by our studies that these decreases are also accompanied by other changes in the materials that offset the beneficial effects of these CTE reductions on thermal shock resistances.

The differences in the nature of the principal four grades of graphite are more clearly apparent in the studies of their mechanical properties as functions of fluence. The brittle ring strength values and their changes as functions of fluence show that the neutron damage up to about 5 × 10^{21} neutrons/cm^2 increases the strengths of grades AXF-5QBG-3 and P-03 by 15 and 30%, respectively, and then the strengths of both decrease with additional fluence. Grade AXF has a steady decrease. All three have 30 to 50% strength decreases in the 30 to 40 × 10^{21} neutrons/cm^2 range. Grade H-395 has a surprisingly different behavior in that its strength continuously increases by as much as 30% at 23 × 10^{21} neutrons/cm^2. Since its CTE values appear to have decreased sharply beyond 10 × 10^{21} neutrons/cm^2 and the change in strain beyond this fluence (discussed below) appears to be saturating, one suspects that the voids caused by irradiation may be smaller and more numerous than those in the other grades. That is — the critical flaw for fracture is remaining small.

The neutron damage appears to have caused immediate and continuous decreases in the fracture strains for all four grades of graphite by 60 to 85% for the terminal values of the fluences of these studies. The actual fracture strains for all at the highest fluences fall within the narrow range of 0.18 to 0.23%. The fracture strains appear to be the most seriously altered property of the four grades of graphite studied.

Young's moduli for grades AXF, AXF-5QBG-3, H-395, add P-03 versus fluence accumulated at 715°C extend these data to >40 × 10^{21} neutrons/cm^2 for isotropic grades of graphite, more than twice that of previous work.[3] The results suggest that in the vicinity of 40 × 10^{21} neutrons/cm^2 that Young's moduli rates of increase with fluence are slowing and/or saturating for grades AXF-5QBG-3 and H-395 for increases of about $\geq$ 300%. One may anticipate that this will happen soon for grade AXF with additional fluence. A different behavior of a parabolic increase and subsequent loss from neutron damage. This is probably a stronger functi the way this grade was fabricated than th nature of the material used in its fabri In it, relatively large particles are bo together with a small volume of porous m The relative smallness of volume of mate apparently tended to "magnify" the closi then generation of porosity by the neutr damage.

Graphite grades AXF and AXF-5QBG be a like manner in that their shear moduli crease at essentially the same linear ra fluence and then, at different maxima of accumulations, decrease parabolically in similar manner. These maxima occur at a 33.8 × 10^{21} neutrons/cm^2 for grades AXF AXF-5QBG. The change in shear moduli fo AXF-5QBG-3 versus fluence would be simil that of grades AXF and AXF-5QBG since th previous properties of grade AXF-5QBG-3 been similar to those of grade AXF. As might expect, the decreases in the chang shear moduli versus fluence following th respective maxima for all four graphite have reasonably direct relationships to neutron-induced volume changes.

Figures of merit (FOM) for thermal resistances calculated as functions of f using the ratios of the fracture strains average CTE values show that the FOM val decrease with fluence for all four grade graphite. The decreases are about 30% f grades AXF and H-395 and 65% for grades and AXF-5QBG-3. The reductions in avera values by neutron damage, as described a were accompanied by unfavorable ratios o harmful changes in the mechanical proper these grades of graphite relative to the shock resistances.

Conclusions

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References

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Graphite grades AXF and AXF-5QBG behave in a like manner in that their shear moduli increase at essentially the same linear rate with fluence and then, at different maxima of fluence accumulations, decrease parabolically in a similar manner. These maxima occur at about 33.8×10^{21} neutrons/cm^2 for grades AXF and AXF-5QBG. The change in shear moduli for grade AXF-5QBG-3 versus fluence would be similar to that of grades AXF and AXF-5QBG since the previous properties of grade AXF-5QBG-3 have been similar to those of grade AXF. As one might expect, the decreases in the changes of shear moduli versus fluence following their respective maxima for all four graphite grades have reasonably direct relationships to their neutron-induced volume changes.

Figures of merit (FOM) for thermal shock resistances calculated as functions of fluences using the ratios of the fracture strains to the average CTE values show that the FOM values decrease with fluence for all four grades of graphite. The decreases are about 30% for grades AXF and H-395 and 65% for grades P-03 and AXF-5QBG-3. The reductions in average CTE values by neutron damage, as described above, were accompanied by unfavorable ratios of harmful changes in the mechanical properties of these grades of graphite relative to thermal shock resistances.

2

Conclusions

The reduction of the CTE values of the high-strength, near-isotropic grade AXF graphite by neutron damage does not improve the calculated thermal shock resistance of this graphite. In fact, the higher fluences are harmful to all of the parameters of the principal four grades of graphite with two exceptions; the CTE of grade P-03 returns to approximately its original value and the strength of grade H-395 is still increasing at a fluence of 23×10^{21} neutrons/cm^2. Otherwise, within the limits of this study, at the higher fluences the volume changes increase at a relatively high rate and the physical properties values decrease.

References

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