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RADIATION EFFECTS IN GRAPHITE IN RELATION TO
ITS ROLE IN FUSION REACTORS^(a)

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SUMMARY

Graphite has several properties which make it a potential material for some CTR components. The response of graphite under irradiation must also be considered in the engineering design stage. The authors review radiation effects data which have been compiled in support of fission reactor designs and briefly discuss the application of this data to CTR designs.

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Synthetic graphite was chosen for use as the neutron moderator material in the first fission reactors primarily because (1) its neutron absorption cross section is very low and the ratio of its neutron scattering cross section to its neutron absorption cross section is very high, (2) it is available in large quantities at relatively low cost, and (3) it is easily formed and machined to the shapes and sizes required for reactor construction. These reasons are still important, but other special properties of synthetic carbons and graphites (such as their high strength-to-weight ratio, increased strength at elevated temperatures and the wide range of available thermal conductivities) are equally important for some applications in advanced reactor designs. In regard to fusion reactors, synthetic carbons and graphites may offer significant advantages over other materials for such applications as (1) the moderation of high energy neutrons for the primary purpose of converting their energy to useful work, (2) reflection of low energy neutrons back into the tritium breeding portion of the blanket, and (3) thermalization of the high energy neutrons so that they can be more readily absorbed before they penetrate into the superconducting magnet region.

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In the process of moderating the high-energy neutrons, carbon atoms are displaced from their lattice sites. Although the subsequent displacement of additional atoms by the primary knockon atoms can be described in the same general terms as in the case of metals, there are basic differences. One of these differences is that the high energy primary knockon atoms of carbon lose much of their energy through electron excitation processes rather than through direct atom-atom collisions; thus, the total number of atoms displaced in graphite does not depend as strongly on neutron energy as is the case for metals. Another important difference is that the resulting radiation-induced changes in dimensions and physical properties are quite unlike the changes which occur in metals. The basic differences between the behavior of metals and of graphite arise because the graphite crystals are extremely anisotropic and because of the complex porous morphology of the polycrystalline graphite bodies.

The hexagonal crystalline form of carbon known as graphite is characterized by strong chemical bonds within the large sheets of hexagonal layer planes, and weak van der Waals forces between the layer planes. When irradiated with high-energy neutrons, this extremely anisotropic structure swells in the c-axis direction (perpendicular to the planes) and shrinks in the a-axis directions (parallel to the planes). This happens because mobile interstitials form clusters between the layer planes while relatively immobile vacancies can, by random juxtaposition, occur in rows within the planes. When the number of vacancies in a row exceeds a certain critical value, they collapse into lines which continue to grow by the capture of additional vacancies at their ends.

The ratio of c-axis growth to a-axis shrinkage is about 10 near room temperature but rapidly decreases to about 3 at 400°C. This ratio approaches, but almost certainly never reaches, 2 at temperatures above 1000°C. Thus, irradiation causes the graphite crystals not only to change shape but also to increase in volume, with the volume increase being most pronounced at the lower temperatures.

The increased volume of the graphite crystallites is not, however, directly reflected in an increased volume for the polycrystalline body. Polycrystalline graphites are manufactured by mixing fine graphitizable carbon particles together with a carbonizable binder, pressing or extruding and heating to temperatures above 2400°C, normally 2800 to 3000°C. Figure 1 shows that the resulting structure contains a good deal of microscopic cracks and pores so that its bulk density is in the range 70-90% of theoretical density. Some of this microporosity is preferentially oriented such that it can accommodate some of the irradiation-induced c-axis expansion. Contrary to what one might expect, therefore, polycrystalline graphites shrink rather than swell under many conditions of temperature and neutron fluence.

Actually, radiation-induced dimensional changes in polycrystalline graphites are a complex function of irradiation temperature, fluence, and morphology of the graphite body. Most graphites swell at irradiation temperatures below about 200°C because the rapid c-axis expansion is not sufficiently accommodated by the oriented microporosity. Above 200°C, the ratio of c-axis expansion to a-axis contraction is reduced and oriented microporosity can

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accommodate enough of the c-axis expansion to produce an initial net shrinkage of the graphite body. Eventually, however, the oriented microporosity becomes filled to such an extent that the graphite ceases to shrink and begins to swell rapidly. Once the oriented microporosity becomes filled, further crystal strain can only be accommodated by fracture and the accompanying generation of porosity. This leads to expansion rates which are greater than those of the individual crystallites, at least at irradiation temperatures below about 1200°C. At higher temperatures, radiation-induced creep can evidently accommodate additional crystal strain thereby preventing porosity generation by internal fracturing. It should be noted that, although the irradiations are normally terminated before the dimensional changes become excessive, linear shrinkages of about 15% in the direction parallel to the extrusion axis and linear expansions of 50% in the direction transverse to the extrusion direction have been measured on some irradiated samples.

Figures 2 and 3 show length changes of typical anisotropic polycrystalline graphites both parallel and perpendicular to the extrusion direction while Figure 4 shows volume changes of one of the same graphites. Note that the onset of expansion occurs most rapidly for irradiation temperatures near 1000°C and that the rate of expansion is also greater than at either higher or lower temperatures. Figure 5 shows volume changes of an improved isotropic graphite. Here, the dimensional changes are much less pronounced; however, as with the anisotropic graphites, the expansion rate is highest for irradiation temperatures near 1000°C. This early and quite rapid expansion near 1000°C is typical of most, if not all, graphites.

PERPENDICULAR

FLUENCE
 $n/cm^2 \times 10^{-22}$ (E 0.18 MeV)

Radiation-induced creep of graphite is also a very important engineering parameter. For example, because the dimensional changes are strongly dependent on temperature and neutron fluence, internal stresses will be generated within the graphite blocks due to the differences in dimensional change rates. In the absence of a stress relief mechanism, these differential stresses would exceed the rupture strength of the graphite and cause the blocks to fracture.

While thermally induced creep rates of graphite are negligible at temperatures below about 2000°C, significant radiation-induced creep has been observed at all temperatures studied (100 to 1200°C). Linear dimensional changes due to creep, ϵ , can be described by the equation

$$\epsilon = A\sigma(1 - e^{-a\phi}) + B\sigma\phi$$

where A and B are transient and steady-state creep coefficients respectively, a is a constant of the order of $1 \times 10^{20} \text{ n/cm}^2$, σ is the applied stress, and ϕ is the high-energy neutron fluence. The transient and steady-state creep are shown schematically in Figure 6. The temperature dependence of the transient creep coefficient, A, is not well known but it is thought to vary only slightly, if at all, with temperature. On the other hand, the steady-state creep coefficient, B, is strongly dependent on temperature as shown in Figure 7.

The steady-state creep coefficients have been shown to be about equal for tensile and compressive stresses and to be constant up to fluences of at least $6 \times 10^{21} \text{ n/cm}^2$. However, Figure 8 shows some recent data up to

CREEP STRAIN →

FAST NEUTRON FLUENCE →

$12 \times 10^{21} \text{ n/cm}^2$, obtained at Battelle-Northwest, which indicates that creep in compression may hasten the onset of expansion; this is a subject that needs to be explored in more detail.

Typical radiation-induced changes in physical properties are shown in Figures 9 and 10 and can be summarized as follows. Young's modulus and strength increase initially but eventually decrease drastically as inter-crystalline cracks begin to form. Thermal conductivity decreases rapidly at first but then saturates. Thermal expansion may decrease, increase, or remain essentially unchanged depending on the type of graphite.

Because the total number of carbon atom displacements per atom (dpa) is not a strong function of neutron energy above 10 MeV, property changes per dpa observed in fission reactor irradiations can, to a first approximation, be used to predict behavior in fusion reactors.

The foregoing summary of radiation effects in graphite reflect nearly 30 years of research which was begun when the first Hanford reactor was built. This work, which was carried on not only at Hanford but elsewhere around the world, has resulted in improved power reactor designs which can operate reliably at increased temperatures and to higher life-time fluences. The basic understanding of radiation-effects phenomena which has been developed and the extensive compilations of data obtained in irradiation testing programs, can furnish a solid foundation for predicting graphite behavior for CTR

applications. It should be realized, however, that the environment to which graphite may be subjected and the requirements for performance of the graphite may be quite different for fusion reactors than for current fission reactors. For example, the optimum temperature-fluence regime may well be different than that of interest for fission reactors. The effects of higher energy neutrons and the effect of differences in atomic displacement rates needs further examination. The effects of such high-energy reactions as

need to be assessed; and radiation-induced creep at high-fluences needs additional investigation.

The importance of obtaining answers to these questions will depend heavily on the design of the fusion reactor and on the role graphite will play in the reactor. On the other hand, at least preliminary answers to some of these questions will be required in order to use graphite to the best advantage. It is essential, therefore, that a close liaison be maintained between design engineers and those scientists conducting research on radiation effects in graphite.

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