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MASTER
DEVELOPMENTS IN SOLID MODERATOR MATERIALS

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1. INTRODUCTION

Work conducted in recent years at several laboratories in the United States has greatly extended the knowledge on a number of solid moderator materials. This review is limited to the three classes of materials on which the most effort has been devoted: carbons and graphite, beryllium oxide, and metallic hydrides. Although a considerable amount of work has been reported on beryllium, this material has not found extensive use as a moderator, and therefore will not be discussed in this paper.

Graphite is used to greatest advantage in large power reactors or in applications in which the moderator is subjected to very high temperatures. Emphasis has, therefore, been placed on long-term radiation effects at high temperatures and on gas - graphite compatibility studies. The importance of graphite as a moderator material is evidenced by the relatively large number of graphite-moderated reactors recently completed or under construction in the United States. These include: Hanford N Reactor; Hallam Sodium-Cooled Reactor; Experimental Gas-Cooled Reactor (EGCR); Peach Bottom High-Temperature Gas-Cooled Reactor (HTGR); Ultra-High-Temperature Reactor Experiment (UMTREX); Molten-Salt Reactor Experiment (MSRE); and the nuclear propulsion reactors (KIWI and NERVA).

Beryllium oxide almost uniquely combines the excellent moderating ability, high-temperature properties, and oxidation resistance required for air-breathing nuclear-propulsion applications [1]. It can also be used to advantage in high-temperature reactors, particularly those having small cores. Recent investigations have been concerned with fabrication methods, properties, and radiation effects. A BeO-moderated reactor, the Experimental Beryllium Oxide Reactor (EBOR) is now under construction in the United States.

Of the hydrogen-containing materials, the metallic hydrides have the highest temperature capabilities and offer definite weight and volume advantages. This review is limited to the most promising of these, yttrium, zirconium and titanium hydrides.

2. GRAPHITE

2-1 Manufacture and Properties

A variety of carbon materials can now be produced with a wide range of properties. Structural properties which may be varied during manufacture, although not completely independently, include crystallinity of the binder and filler phases, particle size, pore size, and anisotropy. These properties will, of course, affect the important engineering properties such as strength, thermal conductivity, permeability, thermal-expansion coefficient, and density. They are also important in determining the magnitude of radiation-induced changes to the properties.

The structural features of different graphites are revealed in micrographs, the quality of which has been enhanced recently by improved metallographic and microscopic techniques [2]. The micrographs in Fig. 1 illustrate several different structures.

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The material in Fig. 1a is a conventional nuclear graphite, made from a "needle coke" [3,4] for the EGCR. The high degree of crystal alignment of this type of coke causes the material to fracture into needle- or flake-shaped particles, whereas particles of less-ordered coke are more nearly cubical or spherical in shape. Needle cokes form a highly crystalline graphite, and alignment of the particles during extrusion produces a high degree of preferred orientation.

The material in Fig. 1b is a so-called "raw-coke" graphite. It was manufactured from an uncalcined (raw) petroleum coke. Whereas in the conventional process the coke is calcined prior to mixing with a coal-tar-pitch binder and then baked and graphitized, in making the raw-coke graphite uncalcined coke is mixed directly with pitch binder and they are then calcined together in the baking process [5]. Indistinct particle boundaries give evidence of the fusing of binder and filler. High strength, isotropy, and uniform structure are imparted to the graphite by this process. As will be noted later, this material shows promise of having good dimensional stability when irradiated at high temperatures.

The material in Fig. 1c was made from coal-tar-pitch binder and a filler composed of equal parts of carbon black and petroleum coke. The areas which appear to be binder or matrix are actually filled to a large extent with the non-crystalline carbon black. Although carbon black is often used to lower permeability and increase strength, it has undesirable effects on dimensional stability [6].

Figure 1d is an example of a conventional petroleum-coke graphite that has been modified by a hot-working process [7]. The increase in density and crystal alignment are quite striking. The effects of irradiation on this material are currently being investigated.

There are many other modifications to standard processing methods, some of which are more subtle than the examples cited above. For example, the effects of irradiation on graphites with the following processing variations are presently being studied: furfuryl-alcohol binder or impregnant for low-permeability applications [8,9]; metallic oxides added to raw materials to promote graphitization [10]; graphites made with a variety of special cokes; and pyrolytic graphite.

The fact that a wide range of pure carbon materials can be made has two important effects. The first is that unless raw materials and processing conditions are carefully controlled, the properties will not be uniform or predictable. Even with the most careful processing, variations within a single batch may be undesirably large for some applications. A detailed study by the Los Alamos Scientific Laboratory [11] provides a good indication of the variation in properties that may occur in a typical commercial grade. It is important to account for such variations in reactor design and in irradiation testing. Progress toward increasing the uniformity has been made by carbon manufacturers in recent years.

Although difficulties are experienced in making a uniform carbon material, the wide variety attainable offers the possibility of tailoring a material with desirable properties for a specific application. This possibility has been exploited in the past, particularly in the manufacture of materials having improved resistance to radiation-induced dimensional changes, and it is the basis for a number of investigations now in progress.

The purity of graphite is no longer of overriding importance because enriched uranium is now available to compensate for reactivity losses from impurities in the graphite. However, the added fuel costs for enrichment must be balanced against the cost of purifying the graphite.

Nuclear graphite of high purity is produced by several manufacturers in a variety of sizes and grades. Currently, unmachined grades cost \$1000 to \$1500 per ton in the United States in several-hundred-ton quantities. Machining adds \$500 to \$1000 per ton. Very large bars can now be produced. For example, the core of the EGCR was assembled from monolithic columns 16 in. in cross section and 20 ft. long [12].

Graphites with low permeability to fission products are required in the High-Temperature Gas-Cooled Reactor in order to keep the primary gas coolant to an acceptably low activity level. In addition to low permeability, these materials must also be dimensionally stable and have good strength and thermal conductivity. Substantial progress has been reported in the development of such graphites.

Recent work includes the development and evaluation of low-permeability sleeves [8,13], fuel-matrix graphite [14], and graphite containing boron for control rods [15].

2-2 Irradiation Effects

Since the 1958 Geneva Conference, the determination of property changes produced by reactor irradiation has probably received more attention in the United States than any other moderator-graphite problem. Although there is generally a need for graphites more resistant to radiation-induced changes, it is sometimes sufficient just to know what changes will occur so that designs can be evolved to accommodate them. However, reactor designers have not yet been able to enjoy such a luxury because, in the present state of reactor development, new reactors are often of advance design for which materials data are not available.

The dimensional stability of graphite in high-temperature reactors has been, perhaps, the property of most interest in the United States in recent years. The problem of internal stresses produced by different rates of contraction across a large block is now receiving more attention. Other properties of interest include radiation-induced creep and changes in thermal conductivity and mechanical properties.

2-2.1 Dimensional Stability - Most moderator graphites are formed by extrusion of a mixture of coke and pitch through a die. This partially aligns the coke particles and gives a preferred orientation to the finished graphite that results in different radiation-induced changes parallel and perpendicular to the axis of extrusion. At the 1958 Geneva Conference it was reported that at 400° C and above, graphite contracts both perpendicular and parallel to the direction of extrusion [16]. Data were available to an exposure of 5×10^{20} neutrons/cm²*. Since that time, the effect of temperature has become much better known, and irradiations have been extended in a few cases [17] to 1.6×10^{22} neutrons/cm², which approaches the expected lifetime exposure of an advanced power reactor. The dimensional changes for EGCR graphite perpendicular to the extrusion axis are shown in Fig. 2 and those parallel to the axis are given in Fig. 3. It will be noted, first of all, that the parallel changes are larger than the perpendicular changes, and also that the parallel rate continues to increase, whereas the perpendicular rate decreases at the higher exposures. A particularly interesting feature of the 800° C perpendicular sample is the rather rapid expansion at about 1.2×10^{22} neutrons/cm².

In the temperature range studied (425 to 800° C) the contraction rate generally decreases with increasing temperature. There is some evidence from other tests [18] that the rate for perpendicular samples again increases above 800° C.

The practical consequences of these changes are two-fold: (1) the dimensional changes themselves must be accommodated in the reactor design to assure that fuel and control-rod channels are not excessively distorted, large gas passages do not form, etc.; and (2) there is some concern that the flux and temperature gradients through a large graphite bar will produce different amounts of contraction, which may, in turn, create excessive internal stresses. The potential problems have been analyzed for the EGCR [12,19]. Stress analyses based on viscoelastic theory have been performed for a number of different bar sizes and geometries [20], and it has been concluded that in some cases, stresses may be generated which exceed the breaking strength. For the case of thick-walled cylinders, the maximum tensile stress can be substantially reduced by dividing the cylinder into sectors. However, as the wall thickness is reduced, sectoring becomes much less effective in reducing stresses.

A number of different graphites having a wide range of structures and properties has been irradiated to learn more about the mechanism by which radiation causes graphite to contract, and thereby to develop better materials. Figures 4 and 5 illustrate the range of contraction rates that have been found for several graphites at about 500° C. The raw-coke graphite has not yet begun to contract. Although some contraction is expected at higher exposures, it shows promise of having good dimensional stability. The KC graphite is, like the EGCR graphite, a highly graphitic anisotropic material made

* All exposures for graphite irradiations refer to neutrons with energies greater than 0.18 Mev.

from a needle coke. It has good dimensional stability in the perpendicular direction, and about average stability in the parallel direction. The low-density graphite (1.3 g/cm^3) shows that low density per se does not result in a high contraction rate. The CSGbF is more isotropic than the KC graphite and was processed at a lower temperature (2450°C instead of the 2800°C or higher normally employed), and contracts at a higher rate.

Normally, small samples, about 0.5 in. in diameter or smaller, are tested. In the case of CSGbF, however, large bars, 4 by 4 by 24 in., have been irradiated and can be compared directly with small samples. As shown in Figs. 4 and 5, they contracted at about twice the rate of the small samples. This size effect, which has also been found for other grades, has been tentatively attributed to the different stress patterns found in small samples and large bars [21].

Although there are undoubtedly several factors that affect the contraction rate, one important factor appears to be the degree of crystallinity of the carbon. Materials that are non-crystalline, either because the raw materials do not graphitize to a high degree or because they have been processed at a low temperature, contract at a high rate. This is illustrated in Fig. 6 by a series of materials, all made from the same petroleum coke but heated to different temperatures [22]. These samples were formed by molding, and dimensional changes were measured parallel to the molding force. In general, the samples heated to the higher temperatures were more stable.

A mechanism for radiation-induced dimensional changes has been proposed that accounts for most of the observations [23,24]. Contraction is attributed to two separable processes. The first is a vestige of the low-temperature damage; it results in an expansion of the crystallites along the c axis and contraction along the a axis. Because this type of damage is annealable, the amount of crystallite expansion and contraction is a function of the irradiation temperature, and this accounts for the temperature dependence of the stability. The second process is non-annealable and is presumed to result from a further densification of the non-crystalline regions associated with grain boundaries and binder junctions. Extensions of this theory have been applied to the qualitative interpretation of dimensional changes in raw-coke graphite, pyrolytic graphite, lampblack-base graphite [6], and the effects of sample size [21] and external stress [25].

2-2.2 Effect of External Stress on Dimensional Stability - Because the graphite moderator is usually constrained from moving or is subjected to an external stress, it is necessary to know how this may affect the dimensional changes during irradiation [26]. The result of moderately low (150 and 300 psi) compressive stresses is to increase the contraction rate. Similar results have been obtained on other graphites for both orientations [25]. In these tests, it was not possible to distinguish between the effects of 150 and 300 psi.

In-reactor creep has recently been measured on a parabolic cantilever beam of graphite for which the extreme fiber stresses are constant along the length [27,28]. The samples were loaded at the end of the beams at several stresses ranging from 475 to 2320 psi. The tests were conducted in a flux of approximately 2.8×10^{13} neutrons/cm²/sec ($E > 0.2 \text{ Mev}$) for 4935 hr (5×10^{20} neutrons/cm²) at 250 to 300°C . The creep rate was apparently enhanced during irradiation, thereby allowing the graphite to sustain greater strain without fracturing. For the specimen under maximum load the total strain was 0.38 per cent, compared with an average out-of-reactor fracture strain of only 0.22 per cent. Because of the importance of understanding the in-reactor creep behavior of graphite, much more work needs to be done to higher doses and over a range of temperatures.

2-3 Gas Reactions

Graphite undergoes thermal- and radiation-induced reactions with many gases, including water, carbon dioxide, oxygen, and hydrogen. The problems in reactor design and operation resulting from these reactions are: (1) a slow reaction of impurities in helium or other inert-gas coolants or gas blankets; and (2) the possible inadvertent exposure of a hot graphite moderator to air by rupture of coolant piping or some similar accident. Each of these problems will be discussed briefly.

2-3.1 Reaction with Trace Impurities - Three helium-cooled, graphite-moderated reactors (EGCR, HTGR and UHTREX) are under construction in the United States, and a fourth, the Hanford N Reactor, utilizes a helium blanket around the graphite. In each of these reactors it is expected that oxidation can be controlled by purging the graphite to remove the initially adsorbed gases and by controlling the amount of impurities in the helium. However, because in most cases the graphite temperatures are sufficiently high to react very rapidly with oxidizing gases, it is important to determine the concentration of each impurity that can be tolerated, the regions over which the moderator oxidation will occur, and the effects on the properties of the graphite and other materials. In addition, the catalytic effects of trace impurities in the graphite must be considered [29,30].

The quantity and composition of adsorbed gases released by heating have been determined for a number of graphites [31]. The gases released up to 1000° C are shown in Figs. 8 and 9. It will be noted that the purest graphites, SP-26-A and AGOT-VCB, evolved much less gas, probably because there are fewer impurity sites capable of holding gases on the surface. The volume of gas released as a function of time at constant temperature was given by the equation $V = A \log t + B$.

2-3.2 Self-Sustained Combustion - The self-sustained combustion of graphite in air is a potential hazard that could arise if the primary coolant system of a gas-cooled, graphite-moderated reactor should accidentally rupture. Because the oxidation of graphite in air is exothermic, it is possible that more heat could be generated than is removed. In such a case, temperatures would be self-sustaining. A number of studies of this condition have been reported for different systems [32-34].

Figure 10 illustrates the conditions which simulate a fuel channel in the EGCR [35]. It is seen that if the temperature is sufficiently high, increasing the flow rate produces more heat by reaction than can be removed. However, the results depend on specific geometries, flow, temperatures, and pressures, and the conditions must be analyzed for each specific reactor design. Several halogen-containing gases inhibit the oxidation of graphite and are being considered as safeguards to be added to the reactor in the event of an accident [36]. However, more development is required to perfect a practical system.

3. BERYLLIUM OXIDE

3-1 Manufacture and Properties

Nuclear-grade beryllium oxide powder is available in the U. S. from a number of commercial sources at approximately \$50 per kilogram. The UOX and Minox AAA grades are probably the best known materials. These powders are usually derived from the sulfate, hydroxide, or basic acetate. Improvements have been made in purity and sinterability in recent years. The total impurity concentration now ranges from a few hundred to several thousand parts per million and usually includes up to 2000 ppm $\text{SO}_4^{=}$ or OH^- and on the order of 100 ppm each of F, Fe, C, Na, and Si. Impurities with high cross section, such as boron and cadmium, have been reduced to less than 1 ppm.

Fabrication methods employed for BeO are the standard ceramic processes [37]: extrusion, cold pressing, hot pressing [38,39], and even hot extrusion [40]. Choice of the method is governed by the size and shape of the desired body. Dimensional control is difficult and finish grinding is usually necessary. Typical costs for material and fabrication vary from approximately \$2 for a 10-cm long by 0.5-cm dia. extruded tube or rod, to \$100 for a cold-pressed 4 by 10 cm brick, ground to a surface flatness of about 0.005 cm over a 10-cm length.

Sintering behavior is a critical factor and remains a practical fabrication problem. It is influenced by the particle size and impurities of the powder and by the temperature, atmosphere, time, and heating rate [39,41-47]. Interaction of these variables is complex and, therefore, sintering characteristics are often a part of the powder specifications. Mixing of powder lots is employed to reduce the lot-to-lot variation, and additives such as MgO, CaO, and bentonite are employed as sintering aids [39,45,46,48]. Sintered bodies 96 to 98 per cent of theoretical density can be produced with satisfactory control of grain size.

The extensive data obtained on properties of BeO in the last few years have been reviewed recently [1]. In general the properties of BeO are characteristic of brittle, ceramic materials with thermal and oxidation stability extending to high temperatures [39,48-54]. The principal problems to be resolved include thermal-stress resistance, corrosion by moisture at elevated temperatures, and radiation stability.

3-2 Irradiation Effects

The radiation stability of BeO has been studied intensively in the U. S. over the last four years. Changes in dimensions and engineering properties as they are affected by composition and microstructure have been emphasized. Currently, glass-phase additives and specimen-size effects are receiving attention.

3-2.1 Dimensional Stability - Dimensional changes have been defined reasonably well for low-temperature irradiations. They result from two processes: lattice expansion, and the formation of grain-boundary cracks. The latter process is a consequence of the anisotropy of the lattice expansion; greater expansion of the c axis develops stresses sufficient to separate the boundaries between grains of unlike orientation. These two processes lead to two approximately linear rates of expansion with dosage, as shown in Fig. 11. The initial rate of 0.2 to 0.3 per cent per 10^{20} nvt* corresponds to the lattice expansion, whereas the subsequent high expansion rate of 0.9 per cent per 10^{20} nvt reflects the additional volume expansion due to grain-boundary cracks. Continued expansion at this higher rate leads to friability and powdering of the materials [55-62].

Variations of the expansion within this general pattern occur with different microstructures. The most important variables are grain size and preferred orientation. The dosage at which the microcracking begins is higher the smaller the grain size; values range from 2×10^{20} nvt for 5- μ material to less than 4×10^{19} nvt for 80- μ material. Preferred orientation of the c axes of grains also tends to increase the dosage at which the microcracking begins. Unfortunately the desired combination of preferred orientation and small grain size has not yet been achieved.

A number of questions regarding expansion at elevated temperature are yet to be resolved. The dimensional changes are progressively smaller the higher the irradiation temperature, and they appear to depend upon both the temperature and the neutron flux [50,61-63]. Changes in specimens of 20- μ grain size irradiated under high-flux conditions (1.5 to 4×10^{14} nv) are illustrated in Fig. 12. The general pattern is thought to be given in terms of the defect production rate, K , and annealing rate during irradiation, A , by the equations:

$$\begin{aligned} \left(\frac{\Delta V}{V}\right) &= \frac{K}{A} [1 - \exp(-At)] \\ \text{where } K &= 3.3 \times 10^{-23} \phi \\ \text{and } A &= 6.3 \times 10^{-5} \exp[-10,400/RT] \end{aligned}$$

$\left(\frac{\Delta V}{V}\right)$ is the volume increase exclusive of that due to grain-boundary separation; ϕ is the neutron flux greater than 1 Mev; R is the molar gas constant in cal/mole/°K; T is the irradiation temperature in °K; and t is the irradiation time in seconds. These equations apply to specimens with grain sizes of $\sim 20\mu$ and describe the experimental data for different fluxes reasonably well up to dosages of $\sim 1.5 \times 10^{21}$ nvt. The equations suggest that grain-boundary cracking can be avoided under the irradiation conditions shown in Fig. 13. Efforts are currently underway to verify these predictions.

Processes that contribute to the elevated-temperature expansion include lattice expansion, grain-boundary separation, defect clusters, and helium bubbles. Above $\sim 700^\circ$ C the lattice expansion occurs primarily in the c axis and becomes smaller as the temperature increases. The expansion caused by grain-boundary separation at elevated temperatures is comparable to that in 100° C irradiations except that it occurs at total expansions that range from 0.4 per cent at 100° C to approximately 1.0 per cent at higher temperatures. In contrast to the low-temperature behavior, the lattice expansion and

*Exposures for BeO irradiations are in units of neutrons/cm² (nvt) with energies > 1 Mev. To convert to neutrons > 0.18 Mev, multiply by 2.

grain-boundary separation do not account for all of the observed volume increase. Helium bubbles and defect clusters probably account for the remainder of the expansion.

The role of helium in the expansion is one of the major questions to be resolved. Helium is formed in neutron reactions with Be at a rate of 0.35 to 0.4 cm³/g BeO per 10²¹ nvt [64] and accumulates in bubbles at the grain boundaries at temperatures as low as 600° C. The bubbles, which are observable in fracture surfaces (Fig. 14), account for 10 to 20 per cent of the expansion of specimens irradiated at 1000° C to 10²¹ nvt. At higher dosages, they are expected to contribute to further expansion.

Less expansion is anticipated in bodies with small dimensions because of the shorter path for helium diffusion and escape. From these considerations, it is estimated that the specimen diameters or cross sections necessary to achieve the results indicated in Fig. 13 will range from 0.3 cm for fluxes of 10¹⁴ nv to ~ 1.3 cm for 2 x 10¹³ nv.

3-2.2 Radiation-induced Property Changes - Property changes depend to a considerable degree on whether or not grain-boundary cracks have formed. Elastic constants exhibit small changes (up to 1.5 per cent) which appear consistent with the change in density. However, once grain-boundary separation occurs, the elastic constants cannot be measured by resonant-frequency techniques.

The modulus-of-rupture strength is increased by irradiation at low doses, followed by a marked decrease which accompanies grain-boundary separation. This trend is shown in Fig. 15 in which the fraction of unirradiated strength is compared to volume expansion. Once the cracking has occurred, the strength changes are somewhat less severe in materials of fine grain size.

Thermal conductivity is currently being investigated. Measurements have confirmed the data presented at the 1958 Geneva Conference [65,66] and have established that the thermal conductivity at 550° C is 60 to 85 per cent of the unirradiated value following exposure to ~ 1.5 x 10²¹ nvt at 550° C to 1000° C.

The effect of irradiation on BeO corrosion by moisture at elevated temperatures has been examined briefly [67,68]; observed corrosion rates under irradiation agreed within 20 per cent with the calculated rates [69,70] and were within the experimental accuracy.

4. METALLIC HYDRIDES

Although a large number of the transition metals form metallic hydrides, factors such as availability, neutron-absorption cross section, and chemical and high-temperature stability limit those useful for nuclear moderators to TiH_x, ZrH_x and YH_x. This review is further restricted to the non-fuel bearing moderators and therefore does not include work on TRIGA and SNAP fuel elements.

Titanium has a relatively high absorption cross section and TiH_x is, therefore, only of marginal interest as a moderator. However, titanium can be readily hydrided in the solid massive state to an N_H^{*} of greater than 9, which is the highest hydrogen concentration of any high-temperature hydrogenous substance [71]. This unique property, which, if not found useful as a moderator, is certainly of first importance for shielding applications [72].

The two hydrides which have been the subject of extensive moderator interest are ZrH_x (N_H = 6.5) and YH_x (N_H = 5.5). Massive forms of both can be prepared directly by equilibration of the solid metal with hydrogen at elevated temperatures. The hydrided metal retains an integral metallic-like structure, having a thermal conductivity at least equivalent to that of the parent metal. A considerable technology exists which defines precise procedures for producing hydride moderators of optimum mechanical and metallurgical properties [73-76].

All hydrides, on being heated to elevated temperatures, decompose into hydrogen and parent metal, and the nature of this decomposition has been studied in considerable detail [77]. The hydrogen content of TiH_x, ZrH_x, and YH_x in equilibrium with one atmosphere of hydrogen is given as a function of temperature in Fig. 16. Such data are particularly useful for gauging the ultimate temperature cap-

* N_H = hydrogen atoms/cm³ x 10⁻²².

abilities of each of these hydrides. The hydrogen partial pressure of these hydrides below 500° C is so low that the use of the unclad hydride is considered practical. It has been established that at high temperatures (500 to 1000° C) containment of a hydrogen pressure of one atmosphere against a zero external hydrogen pressure is practical using metallic cladding [78]. However, it is difficult to contain significantly greater hydrogen pressures. Clad YH_x may be used to 1000° C, and clad ZrH_x to about 700° C.

A considerable amount of information has been obtained on the physical and mechanical properties of the hydrides [75,79,80]. Typically, the hydrides have better thermal conductivity than the parent metals, and although brittle at low temperatures, they exhibit useful ductility at higher temperatures. Their strengths are better than those of the common heat-resistant alloys, enabling them to support structural loads in many applications. The compressive deformation of YH_x ($N_H \approx 5$) is compared with stainless steel in Fig. 17.

The thermal expansion of hydrided zirconium, shown in Fig. 18, closely approximates that of Type 446 stainless steel, whereas hydrided yttrium has a thermal expansion approximating that of the Fe-25Cr-5Al alloy. These heat-resistant alloys are useful cladding materials for each of the hydrides whose thermal expansion they match [78].

The fabrication and evaluation of alloy-clad yttrium hydride have received more attention than clad titanium or zirconium hydrides since this material offers the greatest potential for use at the highest temperatures. Although it has proven difficult to form a metallurgical bond directly to the hydride, an indirect process has been developed which is wholly satisfactory [76]. In the case of yttrium hydride, the metal is first machined to size, allowing for expansion during hydriding, coated with a 1-mil layer of chromium, heated to approximately 1200° C, hydrided, and clad with Fe-Cr-Al alloy by hot isostatic bonding. This treatment produces an integral structure with an excellent metallurgical bond between the heat-resistant alloy and the chromized surfaces of the hydride moderator. Extensive laboratory evaluations of this material in air at temperatures to somewhat above 1000° C have shown excellent geometric stability and hydrogen retentivity for periods in excess of 10,000 hr. In-reactor evaluations have been completed for periods in excess of 8,000 hr without any evidence of adverse radiation effects, loss of hydrogen, or geometrical instability [81].

Long-term evaluation of these materials and related metal-hydride research is continuing at several laboratories in the United States in support of advanced reactor programs.

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FIGURE CAPTIONS

1. a. EGCR Graphite (60X)
b. Raw-coke Graphite (60X)
c. Carbon-black Graphite (60X)
d. ZT-Grade Hot-worked Graphite (60X)
2. Radiation-induced Dimensional Changes of EGCR Graphite. Perpendicular Orientation.
3. Radiation-induced Dimensional Changes of EGCR Graphite. Parallel Orientation.
4. Radiation-induced Contraction of Several Graphites at 500° C. Perpendicular Orientation.
5. Radiation-induced Contraction of Several Graphites at 500°C as a Function of Heat-treatment Temperature.
6. Radiation-induced Contraction of Graphite at 500° C as a Function of Heat-treatment Temperature.
7. Effect of External Stress on Dimensional Changes at 625° C. EGCR Graphite with Perpendicular Orientation.
8. Cumulative Volumes of Nitrogen, Water, and Total Hydrocarbons Evolved by Various Graphites at Temperatures to 1000° C.
9. Cumulative Volumes of Carbon Monoxide, Carbon Dioxide, and Hydrogen Evolved by Various Graphites at Temperatures to 1000° C.
10. Self-Sustained Burning of Speer Nuclear-2 Graphite in Air. Inlet Temperature is 150° C.
11. Components of Volume Expansion in BeO Irradiated at 100° C.
12. Volume Expansion of AOX-Grade BeO Specimens, 20- μ Grain Size and 2.9 g/cm³ Density, as a Function of Exposure and Irradiation Temperature.
13. Photomicrograph of Helium Bubbles in Fracture Surface of UOX Specimen of 15- μ Grain Size and 2.9 g/cm³ Density, Irradiated to $\sim 9 \times 10^{20}$ nvt at 950° C.
14. Estimated Minimum Irradiation Temperature to Avoid Grain-Boundary Separation in BeO of 20- μ Grain Size and 2.9 g/cm³ Density.
15. Strength Changes in AOX-Grade BeO of 20- μ Grain Size and 2.9 g/cm³ Density After Irradiation to Exposures up to 1.5×10^{21} nvt at 100° to 1000° C.
16. Content of Absorbed Hydrogen in Metallic Zirconium, Titanium, and Yttrium in Equilibrium with One Atmosphere Hydrogen at Various Temperatures.
17. Compressive Deformation of Hydrided Yttrium as a Function of Temperature under Load for 100 Hours.
18. Linear Thermal Expansion as a Function of Temperature for Zirconium and Yttrium Hydride and Potential Cladding Materials.

FIGURE 1a. EGCR GRAPHITE (60X)

FIGURE 1b. RAW-COKE GRAPHITE (60X)

FIGURE 1c. CARBON-BLACK GRAPHITE (60X)

FIGURE 1d. ZT-GRADE HOT-WORKED GRAPHITE (60X)

FIGURE 2. RADIATION-INDUCED DIMENSIONAL CHANGES IN EGCR GRAPHITE. PERPENDICULAR ORIENTATION

FIGURE 3. RADIATION-INDUCED DIMENSIONAL CHANGES IN EGCR GRAPHITE. PARALLEL ORIENTATION

NIGHTINGALE, R. E., COLLINS, C. G., and MCGURTY, J. A., Developments in Solid Moderator Materials, Figs. 1-3.

FIGURE 4

RADIATION-INDUCED CONTRACTION OF SEVERAL GRAPHITES AT 500 C. PERPENDICULAR ORIENTATION

FIGURE 6

RADIATION-INDUCED CONTRACTION OF GRAPHITE AT 500 C. AS A FUNCTION OF HEAT-TREATMENT TEMPERATURE

FIGURE 5

RADIATION-INDUCED CONTRACTION OF SEVERAL GRAPHITES AT 500 C. PARALLEL ORIENTATION

FIGURE 7

EFFECT OF EXTERNAL STRESS ON DIMENSIONAL CHANGES AT 625 C. EGR GRAPHITE WITH PERPENDICULAR ORIENTATION

NIGHTINGALE, R. E., COLLINS, C. G., and MCGURTY, J. A., Developments in Solid Moderator Materials, Figs. 4-7.

FIGURE 3

CUMULATIVE VOLUMES OF NITROGEN, WATER, AND TOTAL HYDROCARBONS EVOLVED BY VARIOUS GRAPHITES AT TEMPERATURES TO 1000 C

FIGURE 4

CUMULATIVE VOLUMES OF CARBON MONOXIDE, CARBON DIOXIDE, AND HYDROGEN EVOLVED BY VARIOUS GRAPHITES AT TEMPERATURES TO 1000 C

FIGURE 10

SELF-SUSTAINED BURNING OF SPEER NUCLEAR-2 GRAPHITE IN AIR. INLET TEMPERATURE IS 150 C

FIGURE 11

COMPONENTS OF VOLUME EXPANSION IN BeO IRRADIATED AT 100 C

FIGURE 12

VOLUME EXPANSION OF BeO SPECIMENS, OF APPROXIMATELY 20-MICRON GRAIN SIZE

NIGHTINGALE, R. E., COLLINS, C. G., and MCGURTY, J. A., Developments in Solid Moderator Materials, Figs. 8-12.

FIGURE 13. ESTIMATED MINIMUM IRRADIATION TEMPERATURE TO AVOID GRAIN-BOUNDARY SEPARATION IN BzO OF 20-MICRON GRAIN SIZE AND 2.9 G/CM³ DENSITY

FIGURE 15. STRENGTH CHANGES IN AOX-GRADE BzO OF 20-μ GRAIN SIZE AND 2.9 G/CM³ DENSITY AFTER IRRADIATION TO 1.5×10^{21} NVT AT 100 TO 1000 C

FIGURE 17. COMPRESSIVE DEFORMATION OF HYDRIDED YTTRIUM AS A FUNCTION OF TEMPERATURE UNDER LOAD FOR 100 HOURS

FIGURE 14. PHOTOMICROGRAPH OF HELIUM BUBBLES IN SURFACE OF AOX-GRADE BzO OF 18-MICRON GRAIN SIZE, 2.9 G/CM³ DENSITY, IRRADIATED TO 1.5×10^{21} NVT AT 900 C. 400X

FIGURE 16. CONTENT OF ABSORBED HYDROGEN IN METALLIC ZIRCONIUM, TITANIUM AND YTTRIUM IN EQUILIBRIUM WITH ONE ATMOSPHERE HYDROGEN AT VARIOUS TEMPERATURES

FIGURE 18. LINEAR THERMAL EXPANSION AS A FUNCTION OF TEMPERATURE FOR ZIRCONIUM AND YTTRIUM HYDRIDE AND POTENTIAL CLADDING MATERIALS

NIGHTINGALE, R. E., COLLINS, C. G., and MCGURTY, J. A., Developments in Solid Moderator Materials, Figs. 13-18.