

# MODERATOR GRAPHITE FOR HIGH TEMPERATURE REACTORS

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In a graphite-moderated nuclear reactor, a substantial portion of the fission energy is expended within the moderator. For purposes of economical production of power, feasibility of operating graphite moderators at elevated temperatures giving good thermal efficiency must be considered. The advantages gained by thermal annealing of radiation-induced changes in graphite at high temperatures tend to be offset by the increased probability of chemical reaction with reactor coolants, structural materials, and contaminants. The effect of the evolution of large quantities of adsorbed gases from the graphite under the combined influence of radiation and high temperature is to complicate considerably the problem of excluding nuclear poisons from the extensive graphite pore volume. The probable effectiveness of possible measures to overcome these problems, and their consequences in terms of reactor efficiency are discussed.

## I. RADIATION DAMAGE

One of the most, if not *the* most important area of problems confronting the designer of nuclear reactors of the more radical type surrounds the unknown behavior of the structural materials from which such a reactor must be made. The problems which have already been encountered in the use of materials in more conventional ranges of temperature with more familiar coolants have become fairly well known. These, for the most part, are centered in the phenomena of radiation damage in the solid components and in metallurgical, chemical and radiation chemical effects between materials in contact.

The present paper is concerned specifically with problems centered about the use of graphite as a moderator material in reactors of radical design. Phenomena encountered in its use under conditions found in conventional reactors are being reported in other papers given here, and in recent publications in the British literature. Changes in the mechanical and thermal properties of graphite for integrated flux levels of the magnitude encountered in power reactors are quite substantial. Changes of  $\frac{1}{4}\%$  in physical length

and of a factor of 40 in thermal resistivity certainly cannot be ignored in design.

Fortunately, the problems encountered are less severe than would be anticipated from what has thus far been said. As indicated in Dr. Dienes<sup>1</sup> earlier paper, radiation induced changes in graphite may be largely healed by thermal annealing to temperatures of the order of 500°C. According to abstract, in the following paper Dr. Kosiba<sup>2</sup> will point out that even at irradiation temperatures as low as 100°C, the rate of change in physical expansion with irradiation is cut twofold. It appears, therefore, that by appropriate design, graphite may be used as a moderator in high temperature reactors.

## II. SLOW REACTOR, GENERAL

We turn now to consideration of the general slow neutron reactor. Nuclear-wise, the reactor is comprised of a suspension of U<sup>235</sup> "fuel" atoms in a lattice separated by large numbers of light "moderator" atoms of low absorption cross-section. The nearest ap-

<sup>1</sup> J. J. Antal, R. J. Weiss and G. J. Dienes, This volume, page 137.

<sup>2</sup> W. L. Kosiba, G. J. Dienes and D. H. Gurinsky, This volume, page 143.

proach to this, in principle, is a homogeneous reactor formed by dispersing the fuel within a solid or a liquid comprised predominantly of either hydrogen, deuterium, beryllium or carbon. In a power reactor, the rapid transfer of thermal energy from within the reactor to a working fluid is necessary. This can be done satisfactorily only by a convection process, and hence requires either the circulation of a liquid reactor fluid itself through a heat exchanger, or the introduction of a foreign heat transfer fluid into the reactor core to remove the thermal energy. If such a coolant fluid is used, consideration of the relationship of fluid heat capacity, film heat transfer rates, and feasible fluid velocities demands the use of coolant channels through the reactor core.

But with the introduction of channels for the passage of coolant fluid through the reactor core the reactor becomes no longer strictly homogeneous. To concentrate the fuel material, wherein more than 95 % of the thermal energy is liberated, physically adjacent to the coolant channels is a rather obvious further measure to simplify thermal transfer to the coolant medium. We are then left with a three phase core, comprised of fuel, moderator and coolant fluid. In the interests of neutron efficiency we add a reflector to reduce the number of neutrons lost, a variable slow neutron absorber to permit control and the necessary thermal and biological shields to permit approach of living organisms. We have thus generated a rather general description of a slow neutron power reactor. We focus our attention now on the moderator material.

Superficial thermodynamic considerations tell us immediately that in the interest of efficiency we wish to remove the heat from the reactor fuel at the highest possible coolant temperature. A simple design thus requires that the moderator medium also ride at, or slightly above, this coolant temperature. Common hydrogenous materials are not stable at elevated tem-

peratures, particularly in the presence of radiation. Beryllium is nuclear-wise a preferable material, but economic considerations make carbon more favorable at the present time for use as a moderator material at elevated temperatures.

### III. GRAPHITE AS A MODERATOR

From a nuclear point of view, the closest possible compaction of carbon atoms will "thermalize" the fast neutrons emerging from the fuel in the minimum diffusion distance. A close spacing between fuel-containing coolant tubes can thus be realized with a savings in critical fuel mass required for reactor operation. The most dense aggregate of carbon atoms, diamond, is obviously of no practical concern here, leaving us graphite with an ideal density of about  $2.26 \text{ g/cm}^3$  as the next best choice. In actual practice it is at present feasible to obtain large volumes of pure grade graphite in densities of only about  $1.7 \text{ g/cm}^3$ . This density permits the use of a grid of fuel tubes spread at distances in the range of from 8 to 12 inches, depending upon fuel slug dimensions, fuel enrichment, and coolant nuclear cross-section.

Unfortunately, the raw material from which carbon is obtained for use in the manufacture of artificial graphite contains several elements whose nuclei have a very high cross-section for absorption of the all-important slow neutrons we are seeking to utilize constructively. The most obnoxious of these is boron, with a nuclear cross-section of 750 barns, approximately 200,000 times that of .0032 barn for carbon. It is evident that the presence of a few parts per million of boron as an impurity in the graphite moderator will have a profoundly adverse effect. Fortunately, measures were long ago developed in the manufacture of spectrographic electrodes, whereby residual traces of metallic impurities were converted to volatile halides and distilled away until their concentration was reduced by orders of magnitude.

Stable coolants in present usage at these

elevated temperatures are generally liquids (gases require excessive coolant tube volumes or excessive flow velocities). Most such liquids are of substantially higher nuclear cross-section than the moderator material, and hence their core volume must be maintained at a minimum value. Because of its porous nature, graphite must be protected from high cross-section coolant liquids to prevent its pore volume from being filled by coolant liquid. Such additional stagnant liquid would add a ruinous contribution to the nuclear absorption within the core.

Two measures suggest themselves for protecting the graphite from coolant penetration: the placing of an impervious layer upon the exposed surface of the graphite itself, or the enclosure of building block elements of the graphite within liquid-tight cans, with suitably protected fuel channels. The latter course is at present being investigated with the use of zirconium metal cans in the sodium reactor experiment being built by North American Aviation, Inc. near Los Angeles. The reason for the choice of zirconium is the combination of good structural properties and low nuclear cross-section found in this metal.

However, one of the most severe problems to be confronted in such a can type design is the non-uniform nature of thermal distortions accompanying heat-up of the core from assembly temperature to actual operating temperature. While it is, in principle, possible to match the average thermal expansion characteristics of suitably oriented graphite to that of zirconium in one direction, such a solution would in practice prove prohibitively expensive for a large scale reactor. As an alternate measure, changes in length are being designed around by allowing adequate tolerances at can ends. Of more critical concern are the consequences of the non-uniform distribution of temperatures during start-up or "scram" transients. Insurance against failure from this cause can best be obtained by the prevention of embrittlement

of the metal from which the can is made. For this reason the embrittling gases evolved from the graphite become an important problem; which problem forms the subject of the balance of this paper.

Numerous studies upon graphite have been made by other workers, invariably showing high content of absorbed oxygen, hydrogen and nitrogen. General statements have been made indicating that complete thermal degassing results in the evolution of a NTP volume of gas at least 90% the volume of the graphite. Neutron and gamma radiation will, in time, promote the release of gas atoms from sorption sites. If other sites do not present themselves for recapture of these atoms, gaseous matter is formed outside the graphite.

At temperatures of 500°C and more, zirconium metal readily forms compounds with oxygen, nitrogen, hydrogen, and carbon oxides. These compounds, present at the surface or in the volume of the zirconium, can lead to significant hardening and embrittlement. Efforts are therefore in order to reduce the amount of gas which will be degassed from the graphite. A series of experiments were undertaken directed toward determining the amounts of and character of impurities which can be thermally degassed from commercial grades of graphite. Attempts were made to correlate the gas content with known manufacturing conditions and with subsequent treatment.

Graphite exhibits affinity for gases by virtue of the same mechanism but to a substantially lower extent than activated charcoal. (As was previously pointed out, most of the gases normally absorbed by exposure of graphite to air are absorbed at high temperatures by the zirconium which is protecting the moderator from penetration by the coolant.) The escape of gas from the graphite can be effected thermally, by a process normally assumed to be due to dissociation of chemical bonds between gas atoms and the solid. However, as mentioned in discussion from

the floor yesterday, the presence of helium gas among the products, which can be removed thermally only at high temperatures from specimens of graphite in which it has been previously placed, shows degassing rates are not controlled solely by dissociation of chemical bonds. Since, furthermore, there seems to be no correlation between structure of the temperature spectrum of gas evolution and the gas content, it is inferred that grain and pore size distribution play an important role in graphite sorption.

The volume of gas evolved from two commercial grades of graphite has been measured during inductive heating in a vacuum for 8 minute periods at successively higher temperatures spaced at 100°C intervals. (See figure 1.) A comparison of the volume of gas removed from graphite which has been subjected to reduction in the concentration of metallic impurities through a halide purification process is seen. Evidently a major portion of the gas appearing during thermal outgassing at temperatures below 1000°C in

the former cases resides at impurity centers which are removed by the purification. The first measure to be taken to reduce the seriousness of the gas desorption problem thus appears to be the identical measure desirable from a nuclear standpoint; i.e. increased purity.

The physical changes occurring in graphite during irradiation are accompanied by changes in the pore structure. In general, an increase in the surface area of pores of large radius of curvature takes place, with a compensating decrease in the importance of small sized pores. This phenomenon is accompanied by a change in the gas sorption characteristics which results first in the release of gas from the small pores, then partial readsorption into larger pores. If during this process other strongly absorptive material (such as zirconium) makes contact with the enclosed gas, a net transfer of contamination can occur.

A marked reduction in the degree to which the pore volume is filled with gas during and

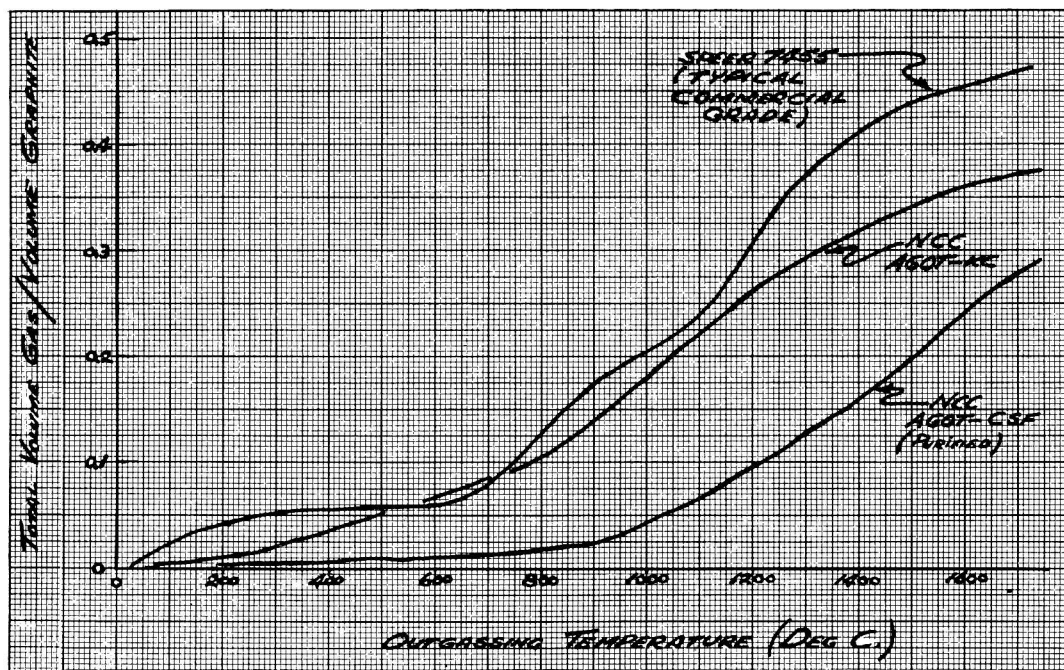


FIG. 1. Gas desorbed vs temperature

after manufacture can be effected by filling the pore volume with an inert gas such as helium or argon. This prevents the pressure gradient accompanying cooling of the graphite from stimulating rapid diffusion of air into the pore volume. The characteristic time for permeation of gas into graphite filled with helium gas is of the order of several months. It is, therefore, of great importance to permit minimum exposure of the graphite between the time of manufacture and installation into the reactor core. This end is achieved by surgical cleanliness in the machining of the graphite to size, by selecting gas resistant wrapping material for

shipment, and by evacuation of the graphite after it has been sealed into its protective container.

Experience with the Sodium Reactor Experiment will serve to guide further developments in design, as well as to provide specific information on compatibility of the component materials under conditions existing within the reactor. It is to be hoped that this paper will point out to industry some of the urgent needs in this rapidly expanding field. Timely research work supported by industry both in industrial laboratories and in universities such as this one, will materially aid the attainment of our goals.