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HAPO GRAPHITE IRRADIATION CAPSULES*

J. W. Helm

INTRODUCTION

Capsules containing reactor-grade and experimental graphites are being irradiated in the ETR, GETR, MTR, and Hanford reactors. These capsules provide two basic types of information about graphite behavior under irradiation. First, information on graphite damage under large neutron exposures equivalent to several years of operation of a graphite-moderated reactor is obtained in a relatively short time. Second, basic information to aid in the explanation of the mechanism of graphite damage under irradiation is obtained. Measurements of many properties are made on each sample in the capsules. These include length, weight, crystallite parameters, electrical resistivity, sonic modulus, thermal conductivity, thermal expansion, etc.

The four basic criteria used in capsule design are achievement of: desired operating conditions, reliability, ease of construction, and ease of disassembly. Two primary irradiation conditions are carefully monitored. These are temperature and neutron flux. Since graphite damage is very sensitive to irradiation temperature, efforts are made to keep the variation below $\pm 10^\circ \text{C}$ at temperatures from 0 to 300°C and below $\pm 25^\circ \text{C}$ above 300°C . Neutron-flux determinations are required so that data can be correlated between various reactors. Consequently detailed studies on neutron spectra are being made for all our graphite irradiation facilities.

*Work performed for the Atomic Energy Commission under Contract AT(45-1)-1350. To be presented at the AEC meeting, "Problems in Irradiation-Capsule Experiments," Germantown, Maryland, October 8-10, 1963.

This report is intended to serve only as a summary of the broad field of graphite irradiation capsules and is written in four parts to serve as a basis for discussion at this meeting. If detailed information is desired the persons listed at the end of each part should be contacted.

Part I summarizes the various capsule designs used for the irradiation of graphite samples. These designs range from highly instrumented capsules in which irradiation temperatures are precisely controlled to capsules in which temperatures are only calculated. In between these extremes are capsules in which the sample temperatures are monitored at several points. All capsule types include monitor materials for measurement of neutron fluxes.

Part II describes several capsule components and materials and the suitability of them for use in irradiation capsules.

Part III outlines the heat-transfer calculations used in the design of the various capsules. The computer program used has proved to be a valuable aid in capsule design and modification.

Part IV presents the methods being used for the calculation of neutron spectra. These spectra are needed to estimate effective activation rates for the monitor materials placed in the irradiation capsules and to correlate radiation data obtained from various irradiation facilities.

PART I. CAPSULE DESIGNS

TEMPERATURE-MONITORED CAPSULE

A typical temperature-monitored capsule for the irradiation of various types of reactor-grade and experimental graphites is shown in Figure 1. In this capsule four quarter-round samples are put together to form a right circular cylinder. They are slid into graphite sample holders, which screw together through the molybdenum centering disks. These disks are fastened onto the aluminum cooling rings. The assembly is slid into the outer aluminum shell, evacuated, filled with helium, and welded shut.

Instrumentation in the capsule consists of five flux-monitors in each cooling ring and at least one thermocouple in each sample position. This capsule provides a relatively simple method for irradiating various graphites. Assembly and disassembly are quite straightforward. Irradiation temperatures from 300 to 1200° C have been achieved with this basic design. Variations on the design include additional graphite cooling disks at the sample midposition, use of six 60 degree samples rather than four quarter-rounds, etc. Sixteen capsules of this basic type have been irradiated successfully to date, and the two others have been partially successful.

GRAPHITE CREEP CAPSULE

An irradiation program for studying the "creep" or deformation of graphite under tensile load is currently underway in the ETR. Sample

temperatures of 475 to 535° C and tensile loads of 600 to 1200 psi are being studied. The heat source is the gamma heating of the capsule components. A schematic of the capsule is shown in Figure 2. One four-inch long graphite sample is positioned between the stainless steel sample grips. The lower grip is held in a fixed position by the heavy aluminum capsule wall. The upper grip is attached by means of a three-foot long stainless steel draw-bar assembly to the bellows. The aluminum heat-transfer block which surrounds the sample is designed with appropriate clearances to provide a flat temperature profile along the sample.

The centerline of the sample is positioned just below the vertical centerline of the reactor so that the entire four-inch sample length is in a region of fairly uniform neutron flux throughout a reactor cycle. A helium atmosphere slightly greater than reactor water pressure is used in the capsule. A differential helium pressure greater than the capsule pressure is controlled on the bellows to provide the desired load on the sample. Sample temperatures are measured by means of seven thermocouples. An unstressed graphite "reference" sample is located in the center hole in the stressed sample to provide a direct measure of the effect of stress.

To date four of these creep capsules have run successfully and another partially successfully. Temperatures from one end of the sample to the other have varied by less than 10° C. Pressure control has been held constant at a given setting plus or minus 2 psi.

TEMPERATURE-CONTROLLED CAPSULE

A capsule for the irradiation of various graphites under controlled temperature conditions is shown in Figure 3. Three or more wafer samples

are put into a sample cup. An assembly made up of the sample cup, one or more pyrolytic graphite insulators, and a platinum heater is held onto the aluminum base plate by a molybdenum spring. The entire assembly is slid into the aluminum shell.

Instrumentation consists of a platinum heater, a thermocouple, and eight flux-monitor wires at each sample position. To make use of all available space in the capsule additional sample sets having only thermocouples are located between the controlled-temperature positions. Sample temperatures have been controlled to plus or minus 10° C at temperatures varying from 600 to 1200° C. One capsule of this type was completely successful and another was partially successful.

BORONATED-GRAPHITE CAPSULES

A rather unique capsule for the irradiation of boronated-graphite samples is shown in Figure 4. Due to the high heating rate from the boron n,α reaction a graphite sample carrier is used to smooth out the temperatures from sample to sample and to provide a heat-transfer path to the aluminum shell. Samples containing up to 8 weight per cent boron have been irradiated successfully in capsules of this type. A helium atmosphere is used in the capsule. Instrumentation consists of several thermocouples and flux monitors.

NON-INSTRUMENTED CAPSULES

A capsule for the irradiation proof-testing of various experimental and prototype graphites is shown in Figure 5. This capsule provides a relatively inexpensive means for direct comparisons of experimental graphites

with a known "reference" graphite. Instrumentation consists of flux monitors. Capsule atmospheres are helium or argon. Various sample configurations are used, from the quarter-rounds shown to hollowed-out graphite boats filled with small cylindrical or parallelepiped samples. Several hundred of these capsules have been run successfully.

FURTHER INFORMATION

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FIGURE 1. TEMPERATURE-MONITORED CAPSULE

IN-REACTOR TENSILE CREEP CAPSULE FOR GRAPHITE

FIGURE 2

E. H. SARGENT & CO.

FIGURE 3. TEMPERATURE-CONTROLLED CAPSULE

FIGURE 4. BORONATED-GRAPHITE CAPSULE

FIGURE 5. NON-INSTRUMENTED CAPSULE

PART II. CAPSULE COMPONENTSTHERMOCOUPLES

Thermocouples are made from bare wire and ceramic insulators by our technicians in our own laboratories.

Wire - Solid 24 gage

Geminol-n and Geminol-p

Compositions:

Geminol-n

96.95% Ni

3% Si

0.05% C

Geminol-p

77.9% Ni

20% Cr

1% Nb

1% Si

0.1% C

Insulators - Two-hole vitreous alumina, 1-in. long.

Some 250 of these thermocouples have been used in capsules irradiated in the ETR, MTR, and GETR. Of these all but 11 have operated satisfactorily. Of the 11 that failed, 5 failed by a break at the right angle bend where the thermocouple enters the graphite sample, 1 failed by a bad junction at the bead, and the other 5 failed at some point along the length of the thermocouple.

Extension Thermocouple Leads. Extension leads are made of duplex Geminol-n, Geminol-p 24 gage solid wire insulated with high temperature polyvinyl.

To date none of the extension leads has failed during operation.

Heaters

Heaters are also made at Hanford. They are flat wafers of platinum wire cast into alundum cement which is fired at 1300° C for 1 hr.

Some 56 of these heaters have been used with varying degrees of success. The main cause of failure has been the reaction of the alundum with the graphite sample cups in the capsule with subsequent shorting of the heater windings.

Connectors

Hermetic Connector. A hermetic connector is used to provide a positive seal for the capsule and a junction for the thermocouples and extension leads. They are leak tight to at least 100 psig helium. In service they are located some 10 ft above the test-reactor core. Both thermocouple and heater leads are exited from the capsule through these connectors. Twenty-four of these connectors have been used with 100 per cent reliability.

RN Connector. The RN connector is a device for making a junction between two pieces of tubing in a reactor vessel. It consists of a six-inch long tube, the I.D. of which is slightly larger than the O.D. of the tubing being joined, and a bored-out tubing union. One piece of tubing is slid three inches into the connector which is then welded to the tubing. The other piece of tubing is slid through the bored-out union and butted up against the first piece of tubing. The union is tightened to complete the junction. The six-inch tube provides a stiffener so that the union does not vibrate loose during reactor operation. The device has been used successfully on nine instrumented capsules in the ETR.

Flexible Tubing

Corrugated stainless steel, one wire braid corrugated flexible metal tubing is used for leadout tubing for our GETR capsules. Thermocouple leads are carried from two feet inside the pressure vessel to the recorder in this tubing.

Bellows

Bellows are two-ply 321 stainless steel, 0.006-inch thick with welded end fittings. The bellows have performed completely satisfactorily in five irradiation capsules operating at differential pressures of 200 psi.

Pressure Controller

The pressure controllers used to control both the capsule pressure and the bellows pressure in the graphite creep capsule are automatically operated, remotely-controlled, pneumatic pressure regulators. They are rated for 0-3000 psi fluid. They have operated satisfactorily for a year of nearly continuous service.

Materials

Aluminum. One problem that comes up from time to time is porosity in wrought aluminum bars. Recently a 20-foot long bar was found to have gas pockets up to 1/8-inch diameter and 1/8-inch long at various locations along its length.

Molybdenum. Molybdenum has proven to be a useful construction material for capsule parts. It has been used to good advantage as a means of limiting heat transfer, supporting graphite sample holders, and as a spring for holding wafer assemblies together.

Pyrolytic Graphite. Pyrolytic graphite serves both as an excellent insulator and an excellent conductor due to its high degree of anisotropy. This makes it a very useful material for capsule construction, particularly at high temperatures. Along the layer planes its conductivity is about the same as tungsten and against the planes it is a better insulator than zirconia. It also has high tensile strength which increases with increasing temperature.

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PART III. HEAT TRANSFER CALCULATIONS

Use of an IBM 7090 computer has greatly facilitated the design of graphite irradiation capsules at Hanford. The temperatures of all capsule parts are now calculated in considerable detail by means of steady-state heat-transfer calculations. The basic computer program used is STHTP - A Steady-State Heat-Transfer Program for the IBM 7090 Computer, HW-73668. The program has the capability of handling three-dimensional geometries involving internal heat generation, constant or temperature dependent thermal conductivity, constant contact or film coefficients, and heat transfer by conduction and radiation. The program has the capacity to handle the following:

- 500 nodes, or regions where heat is generated or transferred.
- 20 sink, or boundary temperatures.
- 30 material-property tables.
- 20 contact or film coefficients.

Any number of heat-generation rates can be used simply by changing the material-property tables and adding a new case card for a new set of calculations.

The technique used by the program consists of an iterative solution of the heat transfer through all the nodes in the problem. An acceptable solution is obtained when the error in heat flux across any node boundary is less than an arbitrary value set up for each capsule. Output from the program consists of:

Temperature at each node.
 Conductance through each of the eight sides of each node.
 Temperature of and heat transferred to each sink.
 Total heat generation in the capsule.

Calculation Details

Nomenclature

A heat-transfer area, or area of node face,
 U thermal conductance,
 T temperature,
 Q heat-generation rate,
 ϵ_N heat-balance error for node N,
 ϵ_T absolute value of heat-transfer error,
 α acceleration factor.

Subscripts

N node number,
 K number of node adjacent to node N,
 J number of node side.

The capsule is divided into as many nodes as are required to provide the amount of detailed temperature distribution desired. The sink or boundary conditions are established and the material-property tables made up.

The temperature of any node, N, can be found in terms of the node heat-generation rate, Q_N , and the adjacent node temperature, T_K , from the following equation:

$$T_N = \frac{Q_N + \sum_{J=1}^8 A_{N,J} U_{N,J} (T_K)}{\sum A_{N,J} U_{N,J}}$$

The computer solves a set of N equations of this type by an iterative process. After a prescribed number of iterations the accuracy of the calculated temperatures is checked by making heat balances on each node. This heat-balance error is given by the equation:

$$\epsilon_N = Q_N + \sum_{J=1}^8 U_{N,J} A_{N,J} (T_K - T_N)$$

Then $\epsilon_T = \sum_{N=1}^N \epsilon_N$ = the absolute value of ϵ_N .

If ϵ_T is less than the prescribed value set up for the capsule, the temperatures are considered accurate and the iteration procedure is terminated.

To speed up the iterative process the program uses an acceleration factor, α . The calculated node temperature, T_N , is compared with the calculated temperature from the previous iteration, T'_N or

$$\Delta T_N = T_N - T'_N$$

A corrected node temperature is then calculated from $T_N = T'_N + \alpha \Delta T_N$. An extrapolation procedure is also used to eliminate some iterations and bring the calculated temperature asymptotically to the correct temperature. Ratios of the ΔT_N are taken for the four previous iterations and used for the extrapolation. A history of the calculated node temperature is shown in Figure 1.

Use

The heat transfer program has been used at Hanford to calculate temperatures in new capsules, provide a basis for design changes in capsules,

test new capsule materials, etc. The basic program has been modified to readily handle the radial symmetry of our graphite irradiation capsules. The areas, A, and distances, d, needed for the computer input for a typical node in radial symmetry are shown on Figure 2.

A typical nodal structure for a temperature-monitored capsule is shown in Figure 3. It is recommended that anyone using such a computer code initially establish the maximum number of nodes that might possibly be of interest even in future calculations. It is much easier to eliminate nodes in later capsules than to completely revise the nodal structure if more detailed temperatures are required.

The results of a typical temperature-monitored capsule calculation are shown in Figure 4. The three lines for each gamma heating rate correspond to concentric graphite node temperatures. The temperature distribution agrees within plus or minus 25° C of the temperatures as measured by thermocouples in irradiation capsules.

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FIGURE 1. CALCULATED NODE TEMPERATURE

Typical Node in Radial Symmetry

FIGURE 2

HEAT TRANSFER NODE DIAGRAM OF A TYPICAL CAPSULE

FIGURE 3

GETR CAPSULE 5

FIGURE 4

PART IV. NEUTRON-SPECTRA CALCULATIONS

Studies are being made on reactor neutron spectra to aid in better understanding the radiation damage to graphite as measured by activation monitors placed in the irradiation capsules. These spectra are required to estimate effective activation rates for the monitors in various reactor positions, to estimate the differences in flux between the monitors and the actual samples being irradiated, and to correlate radiation data obtained in different irradiation facilities in various reactors. Calculations of spectra are being made for graphite capsule positions in the GETR, ETR, and Hanford reactors.

Four different reactor-physics computer codes have been used to calculate fast-neutron spectra: GNU-II, HFN, Program S, and 2DXY. Significant differences in spectra are found with the different codes. Consequently two multi-group, multi-region, transport-theory codes, Program S and 2DXY, are being used due to their greater accuracy and wider applicability. In these studies the Program S code, a one-dimensional code, is set up with 33 energy groups of which 16 are greater than 0.183 Mev and the 2DXY code, a two-dimensional code, is set up with 18 energy groups 16 of which are greater than 0.183 Mev.

The results of the computations give the neutron flux as a function of space and energy. The computations can be readily modified for changes in materials and geometry. The spatial array over the cross-sectional area of

the reactor can be varied. In the case of the GETR the entire core cross-section is calculated using one-inch by one-inch increments. Calculations are in progress to determine the effect of large changes in materials and/or geometry on the spectra in our graphite capsule positions. Such things as replacement of fuel with poison, replacing graphite with cobalt, etc. are being calculated. To date these types of changes have not had significant effect on the spectra; however, flux intensity has been altered to some extent.

The results of the spectra calculations for various locations in the K-Reactor lattice at Hanford are shown in Figure 1. The lattice is a square symmetric lattice with an annular fuel element, water coolant, and graphite moderator. The moderator boundary is the inner edge of the graphite channel and the cell boundary is the midpoint in the graphite between tube channels. The flux distribution, $\phi(u)$, is plotted as a function of lethargy, u , which is defined as $u = -\ln E/E_0$ where E_0 is 10 Mev. It should be noted that the fission spectrum is not a good approximation of the spectra at any position including the center of the fuel.

The marked difference in fast-neutron spectra between a graphite-moderated reactor, C-Reactor at Hanford, and a testing reactor, the ETR, is shown in Figure 2. The C Reactor spectrum shown is for the cell boundary in the graphite moderator and the ETR spectrum is for the center of the G-7 Hanford loop. The two curves have been normalized so that there are equal areas under the curve to lethargy 4.

The large differences in flux intensity which may exist in even the same reactor facility are shown in Figure 3. These spectra are all for the

G-7 Hanford loop in the ETR. The spectra are for one-inch by one-inch area increments in the south third of the G-7 reactor position. The bottom curve, G7-SW, corresponds to the center of the loop with stainless steel specimens and a stainless steel grid work, the middle curve, G7-S, to stainless steel specimens, and the top curve, G7-SE, to the stainless steel pressure tube and the aluminum shroud tube. Thus two positions in the loop only two inches apart have flux intensities differing by a factor of three over the range of neutron energies which produce approximately 90 per cent of the damage to structural materials.

The spectra obtained in these calculations are used to compute effective cross-sections for such fast-neutron monitors as Nickel-58 and Iron-54. The effective cross section, $\bar{\sigma}$, is given by:

$$\bar{\sigma} = \frac{\int_{\infty}^0 \phi(u) \sigma(u) du}{\int_{u_1}^0 \phi(u) du} = \frac{\int_0^{10} \phi(E) \sigma(E) dE}{\int_{E_1}^{10} \phi(E) dE}$$

where $\phi(u)$ is the flux distribution as a function of lethargy and $\sigma(u)$ is the lethargy-dependent activation cross section. Typical differential curves used to obtain $\bar{\sigma}$ for nickel are shown in Figure 4. Since the curves are plotted as a function of lethargy the flux in any lethargy interval is proportional to the area under the curve.

The effective cross section for the energy range chosen to correlate observed property changes is used to calculate neutron exposures from the

monitor activities. For those monitors with high threshold energies, which include most of the commonly used monitors such as sulfur, aluminum, nickel, and iron, the effective cross sections vary considerably throughout the reactor lattice due to changes in the spectra below the threshold energy. Consequently effective cross-section calculations for the energy range of interest must be made for each monitor since a large number of the neutrons damaging the samples may have energies well below the monitor energy threshold. Effective cross sections are especially necessary if comparisons of damage in different spectra in different reactors are being made. Although the monitor may be activated by neutrons having energies in excess of 4 Mev it may be necessary to calculate neutron exposures to samples to neutron energies as low as 0.05 Mev in order to correlate observed property changes.

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FIGURE 1. FLUX SPECTRA-GRAPHITE MODERATED REACTOR

FLUX SPECTRA IN ETR TEST FACILITY AND A GRAPHITE MODERATED REACTOR

FIGURE 2

FLUX SPECTRA AND RELATIVE INTENSITIES IN ETR WATER LOOP

FIGURE 3

FIGURE 4. EFFECTIVE CROSS SECTION FOR NICKEL