DESIGN OF A CAPSULE
FOR IRRADIATING GRAPHITE
AT TEMPERATURES UP TO 2000°C

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SUMMARY

A non-instrumented capsule has been designed for irradiating graphite samples at temperatures up to 2000°C, and a device for measuring the temperature has been designed and laboratory tested. Tungsten heater elements used to help achieve the high temperatures were found to react with the graphite even though there was no direct contact. Despite this reaction, laboratory tests indicate that the tungsten heater elements will be suitable for irradiations up to three or four weeks in duration. A description of the materials and properties to be tested for fusion reactor applications is also given.

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INTRODUCTION

Carbons and graphites have potential for use as curtains or liners that would be located between the plasma and the vacuum wall of fusion reactors.⁽¹⁾ Some reactor design concepts include thick sections (several cm) of graphite in at least a portion of this region, while other designs call for the use of a thin liner which could be either a coating on the vacuum wall or a separate liner that stands between the plasma and the vacuum wall. A thin liner or coating would reduce contamination of the plasma by high-Z ions vaporized or sputtered from the vacuum wall. Thick liners would reduce plasma contamination and also soften the neutron spectrum thereby reducing the helium and hydrogen production rates and displacement damage to the vacuum wall. Thickness of the liner is apparently limited only by heat generation considerations because the temperature must be kept below 2000°C to avoid excessive carbon vapor pressure. Even 2000°C poses a problem, however, in that neutron radiation damage effects have never been investigated at such high temperatures. A great deal of data have been generated on the radiation behavior of so-called nuclear grade graphites at temperatures up to 1200°C, and there are some data up to 1450°C.⁽²⁾ One irradiation test has been performed at temperatures up to 1900°C⁽³⁾, but it was only a single irradiation, and maximum fluences were only $\sim 6 \times 10^{20}/\text{cm}^2$ ($E > 0.18$ MeV). Fluences at least 20 times that high are required to fully establish the radiation behavior relative to that found at lower temperatures. The experiment described in this paper was designed to generate the required data and will be essentially the first series of irradiation tests into this unexplored high-temperature region.

In addition to their possible use as liners, carbons and graphites also have potential for use as limiters, beam stops, and wall armor.⁽¹⁾ These applications likely would require the use of thermal shock resistant materials such as carbon/carbon composites, which have never been

radiation tested at any temperature. It was therefore decided to include some of these materials in the testing program.

This document describes the irradiation capsule that has been designed and briefly discusses some other aspects of this exploratory radiation testing program.

CAPSULE DESIGN

HARDWARE

Figures 1 and 2 show assembled views of the capsule designed for irradiating graphite at 1500° and 1900°C in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL). The top and bottom subassemblies are designed to operate at 1500°C while the two middle subassemblies are designed to operate at 1900°C. Each subassembly is composed of a tungsten tube with tungsten end caps welded in place. The tungsten serves as a heater to achieve the high temperatures. The samples are located inside a graphite tube fitted with graphite end plugs. This graphite assembly is separated from the tungsten both axially and radially with zirconium carbide spacers to minimize the reaction between carbon and tungsten.

The tungsten subassemblies are hermetically sealed to prevent carbon from being transported out of the subassembly and deposited on the surface of the aluminum cladding. Past experience with graphite irradiation testing has shown that this can happen even at temperatures much below those expected in this test. The emissivity of surfaces thus blackened is greatly increased which increases the amount of heat lost via radiation. Changes in emissivity of the surfaces in this experiment involving such high temperatures would change the temperatures substantially as discussed in a later section.

To minimize the amount of oxygen and other impurities within the tungsten subassembly, the whole subassembly will be degassed for 30 minutes in a vacuum of less than 10^{-6} torr at 2000°C and then backfilled with high purity helium prior to making the final closure weld.

To check out the possibility of interaction between the carbon and tungsten, a subassembly similar to those shown in Figure 2 was constructed and laboratory tested. After the subassembly was evacuated, backfilled with helium, and welded shut, it was then maintained in a high purity helium atmosphere at 2000°C for 10 days. This caused very little change in appearance of the tungsten, but it became very brittle. Auger analysis of a fracture surface showed that about 25 at% carbon was present throughout the bulk of the tungsten. A second mock-up subassembly was tested at 1900°C for 21 days. Again, the appearance was little changed, but the tungsten was very brittle, and Auger analysis of a fracture surface showed about 50 at% carbon throughout the bulk. Despite the large carbon content, however, the tungsten swelled only about 1% in each case. There was never any evidence for a reaction between the ZrC and either the carbon or tungsten. The same holds for the ZrC and TiC coatings discussed later.

The mechanism of carbon transport to the tungsten tube can only be speculated; most likely it is simply via carbon vapor. Oxides of carbon, CO and CO₂, remain a possibility although it seems unlikely because of the concerted effort to remove oxygen and other impurities via thorough degassing. Direct contact was definitely eliminated by the ZrC spacers.

In an effort to eliminate or substantially reduce this diffusion of carbon into the tungsten, some tungsten tubes were purchased that were coated on the inside with a thin layer (15 to 25 μ m) of either ZrC or TiC. Subassemblies similar to those shown in Figure 2 were prepared using tubes with both types of coating. These were thoroughly degassed and then heated for one month at 2000°C in a pure helium atmosphere. Again, the appearance changed little but the tubes became very brittle. This time Auger analyses showed 25 to 30 at% carbon throughout. Swelling of the tubes ranged from 1.6 to 2.2%.

Obviously the ZrC and TiC coatings helped but not as much as was hoped. Thicker coatings were judged unacceptable because of the uncertainties that would result in the temperature calculations if the coating thickness were more than about 10% of the tungsten wall thickness. Uncertainties would occur because the gamma heating rates for

the coating materials are less well known than for tungsten. It was also felt that a different method of sealing the subassemblies would be required because the coatings would make it impossible to weld end caps in place.

One brief test was run with tantalum in place of tungsten, but the tantalum was found to pick up just as much carbon as the tungsten. Consequently, a decision has tentatively been made to use the uncoated tungsten tubes for the first one-cycle (~ 23 days) irradiation in HFIR. If the tungsten merely becomes embrittled and swells only slightly but does not fracture or crack, it will be acceptable.

CALCULATED TEMPERATURES

Details of the temperature calculation are given in Appendix I. Figures 3 and 4 show axial and radial temperature profiles, respectively. Temperature gradients in both directions are quite small within the sample areas. The relatively flat axial temperature profiles within the different temperature regions are achieved by varying the wall thickness of the tungsten tubes to compensate for variations in the gamma heating rate with axial position. The very flat axial profile near the upper end of the 1500°C tungsten tube is due to the larger end cap in that position which provides additional heat. Because there is only a small radial temperature gradient between the graphite and tungsten, the size of the helium gas gap and also the emissivities of the graphite and inner tungsten surfaces are relatively unimportant. Thus, uncertainties in only three parameters have significant effects on calculated temperatures: 1) size of the helium gas gap between the tungsten and aluminum; 2) emissivities of the outer tungsten surface and the inside surface of the aluminum tube; and 3) total gamma heating rates and their variation with time during the reactor cycle. Figures 5-7 show temperature variations with these three parameters, respectively.

Maintenance of the gas gap within 1 to 2 mils (0.001 to 0.002 in.) of the design value should be possible thus providing for temperature uncertainties of less than $\pm 20^\circ\text{C}$ from this source. Emissivities of

Figure 3. Axial Temperature Profile

Figure 4. Radial Temperature Profile

Figure 5. Dependence of Temperature on Size of the Helium Gas Gap

Figure 6. Dependence of Temperature on Emissivity of Aluminum Tube

Figure 7. Dependence of Temperature on Gamma Heating Rate

tungsten are fairly well known and are shown in the appendix. However, total hemispherical emissivities for aluminum alloys are reported in the range 0.05 to 0.15. A value of 0.10 was assumed, and Figure 6 shows that temperature can vary $\sim \pm 50^\circ\text{C}$ over the above range of emissivities at the higher temperature. Thus, the major source of temperature uncertainty appears to be uncertainties in the gamma heating rates. Figure 7 shows that uncertainties of just $\pm 10\%$, which is about as well as the gamma heating rates are known, would produce temperature uncertainties of $\pm 100^\circ\text{C}$. Other uncertainties would be superimposed. Thus, total uncertainties in the calculated temperatures of $\pm 200^\circ\text{C}$, or even somewhat larger, seem possible. Thermocouples cannot be used in the reactor position for which this test is designed, but some type of a temperature measurement would be highly desirable.

TEMPERATURE MEASUREMENT

Figure 8 schematically shows a temperature measuring device that was developed and demonstrated in laboratory tests. It is quite simple in principle being based on two materials with quite different thermal expansion coefficients. However, severe restraints are imposed by the in-reactor environment and the temperatures anticipated. The high temperatures and associated compatibility considerations restrict the number of materials that might be used. Further restrictions are imposed by the gamma heating which, to a first approximation, tends to be proportional to density. Thus, use of a high-density material to monitor the temperature of a low-density test specimen suffers from the tendency for the monitor to run hotter than the test specimen. The temperature difference between the two can be minimized by good thermal contact, but this exacerbates the compatibility problem.

Obviously, graphite itself would be ideal as one of the temperature monitor materials in the present case; ZrC was found to be satisfactory as the second. ZrC has a thermal expansion coefficient of about $10 \times 10^{-6}/^\circ\text{C}$ while that for the type of graphite tested was about $1 \times 10^{-6}/^\circ\text{C}$ in the axial direction. The moderately high density of ZrC would result in a rather large in-reactor heat generation rate, but its temperature can easily be kept down to that of the graphite by keeping the ZrC pellet

small and in good thermal contact with the graphite. It was already mentioned that graphite and ZrC are quite compatible at least up to 2000°C.

Figure 8a shows the configuration of the temperature indicator prior to placing it in a reactor or furnace. Figure 8b shows the configuration after it has been heated to some maximum temperature, $T_{\max}$, and then cooled to room temperature again. Since the ZrC expands more than graphite, it forces the tight-fitting graphite plug a short distance out of the graphite holder. Note that the plug and holder are both made out of the same material so that they expand exactly the same. After irradiation (or a furnace run) the indicator can be placed in a dilatometer and its length plotted vs. temperature. A slope change occurs at $T_{\max}$. Alternatively, the overall length, or the step height between the plug and holder, can be measured at room temperature after a series of successively higher heat treatments in a furnace. Until the heat treatment temperature exceeds $T_{\max}$, the step height will remain the same. At higher heat treatment temperatures, the step height will begin to increase. Since the difference in thermal expansion coefficient between the ZrC and graphite is about $9 \times 10^{-6}/^{\circ}\text{C}$, a 1/4 inch long ZrC pellet would result in a step height change of 2.25×10^{-6} in/ $^{\circ}\text{C}$. Laboratory tests have shown that $T_{\max}$ can be determined to within about $\pm 25^{\circ}\text{C}$ using this device.

It remains to be shown how well this temperature indicator would perform during in-reactor tests. Certainly radiation would cause property changes including changes in the thermal expansion coefficients of both materials. It would also cause dimensional changes, particularly in the graphite where the change would be a shrinkage under most conditions. However, with the possible exception of the radiation-induced graphite shrinkage, none of these changes should affect the ability to determine $T_{\max}$. Shrinkage of the graphite would cause the plug to be pushed farther and farther out of the holder. Post-irradiation testing then would tend to yield the most recent temperature rather than $T_{\max}$. Detailed analyses have not been performed, but it seems that extreme and unlikely variations

GRAPHITE
ZIRCONIUM CARBIDE

Figure 8. Schematic Diagram of Device for Determination of Maximum Temperature

in reactor operating conditions would have to occur before $T_{\max}$ would be very different from the most recent temperature.

MATERIALS AND PROPERTIES TO BE TESTED

The types of materials to be tested and the rationale for their inclusion in this test is summarized in the following table.

<u>Materials</u>	<u>Goals</u>
Nuclear Graphites (two types)	Determine lifetimes relative to that at lower temperatures.
Other Bulk Graphites (two types)	First radiation data on graphites with good thermal shock resistance.
Carbon/Carbon Composites (two types)	First radiation data on composites (composites have excellent thermal shock resistance).
Fibers (several types)	Baseline data for same type of fibers used in composites. Extension of data obtained at 470°C.

The primary purpose of this experiment is to compare the radiation behavior of graphite at very high temperatures with that observed at 1200°C and below. To make this comparison, the two types of nuclear grade graphite to be irradiated are ones for which the most data at lower temperatures are available.

Recently there has been increasing interest in the possibility of using graphite for limiters and wall armor. Limiter applications, in particular, would subject the graphite to very high thermal shock loads, much higher than typical nuclear grades can withstand. Thus, the irradiation will include two types of bulk graphite that have good thermal shock resistance and two types of carbon/carbon composites which have even better shock resistance. Virtually no radiation data exist for the latter materials. Finally, individual bundles of fibers, including the same types used in the composites, will be part of this test. Other

types of fibers will include the same types irradiated in an earlier test at 470°C⁽⁴⁾ and thus provide a comparison at the different temperatures. The pre- and post-irradiation measurements planned for these materials are given below:

	<u>Bulk Graphites</u>	<u>Composites</u>	<u>Fibers</u>
Dimensions	X	X	X
Thermal Expansion	X		
Thermal Conductivity (one sample only)	X		
Elastic Modulus	X		
Lattice Parameters (C-axis only)	X		
Electrical Resistivity	X		
Surface Area	X	X	

FUTURE WORK

The work done thus far shows that graphite temperatures up to 2000°C can readily be achieved in the target rod positions of the HFIR despite the small diameter (0.65 in) of the test facility, providing a refractory metal (such as tungsten or tantalum) is used as a heater. However, no way was found to prevent carbon from reacting with the tungsten (or tantalum) and causing its embrittlement. Despite the embrittlement problem, it is felt that the tungsten would survive at least short term (up to one month) irradiation. Plans, therefore, were to begin the first one-cycle (~ 23 days) irradiation during the second quarter of FY-1980. However, funding limitations in the Magnetic Fusion Program have caused an indefinite postponement.

When funding improves, it is recommended that the one-cycle irradiation be conducted as planned. In addition, long-term (up to six months) graphite/tungsten compatibility tests should be started in the laboratory. The results of both the in-reactor test and the long-term laboratory tests must be evaluated before long-term irradiations are attempted.

An alternate approach might be to choose a larger diameter (say two inches or larger) reactor test facility. This would allow more space for the required temperature gradient. The use of tungsten would probably still be required, but it might be possible to operate it at a lower temperature. Drawbacks to this approach are that gamma heating rates and neutron fluxes in available large-diameter reactor test facilities are probably much lower than in the small test positions in HFIR. Also, large test positions normally cost much more.

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APPENDIX A

TEMPERATURE CALCULATIONS

Figure A1 is a schematic diagram showing some of the input parameters used for calculating temperatures via the HEATING 5 computer program.⁽⁵⁾ Other input parameters are given in Table A1 and Figures A2-A4. The capsule was assumed to be symmetric about its axis and about the reactor horizontal midplane.

A capsule surface temperature of 175°C was used based on a maximum heat flux of 308 W/in². This was estimated from a situation where a similar capsule with a heat flux of 381 W/in² was calculated to have an outer surface temperature of 189°C.

The gamma heating rate for graphite is given by Figure A4. Tungsten heating rates were taken to be 1.8 times that. Heating rates for each of the other solid materials were assumed to be the same as for graphite.

To facilitate data input to the computer, gamma heating rates from Figure A4 were fitted to a third degree polynomial; from Z=3 to Z=10, the variation in heating rates was assumed to be linear. These heating rates were then modified to account for the taper of the tungsten tubes.

In addition to conduction through the helium gas in regions 14, 15, 25, and 26, simple radiation heat transfer was included. Emissivities of graphite and aluminum were assumed to be 0.8 and 0.1 respectively; that of tungsten is given in Figure A3. Net emissivities between opposing surfaces are given by

$$\epsilon = \left(\frac{1}{\epsilon_1} + \frac{1}{\epsilon_2} - 1 \right)^{-1}$$

Calculated temperatures are only very slightly dependent on the choice of emissivities inside the tungsten tube and on the size of the gas gap between the graphite and tungsten. Likewise, heating rates used for aluminum and alumina are relatively unimportant.

Figure A1. Input Parameters Used in Temperature Calculations

Table AI. Materials and Heat Generation Rates Used in Heat Flux Calculations

<u>Region(s)</u>	<u>Material</u>	<u>Heat Generation Rate (W/in³)</u>
1,2	ZrC	3100
3	Tungsten	19200
4,10	Graphite	$940 + 0.567(Z) - 8.74(Z)^2 + 0.462(Z)^3$
5	Tungsten	16040
6,7,8	ZrC	2550
9	Tungsten	15325
11	Tungsten	10825
12,13	Al ₂ O ₃	1214
14,15, 25,26	Helium	Zero
16	Tungsten	22588
17	Tungsten	$18646 + 953(Z) - 173(Z)^2 + 0.402(Z)^3 + 0.462(Z)^4$
18	Tungsten	$14260 + 2406(Z) - 151(Z)^2$
19	Tungsten	18870
20	Tungsten	18029
21	Tungsten	$12400 - 604(Z)$
22	Tungsten	$-47120 + 12213(Z) - 483(Z)^2$
23	Tungsten	$3543 + 1953(Z) - 104(Z)^2$
24	Tungsten	12178
27	Aluminum	1214

Figure A2. Thermal Conductivity of Helium

Figure A3. Total Hemispherical Emissivity of Tungsten

Figure A4. Heating Rate for Graphite in Target Rod Positions of HFIR

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