Fuel Performance Improvement Program

The Development of Graphite-Coated Cladding for Improved PCI Performance of LWR Fuels

June 1981

EXXON NUCLEAR
COMPANY, Inc.

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The Development of Graphite-Coated Cladding for Improved PCI Performance of LWR Fuels

June 1981

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ABSTRACT

One of the fuel concepts being tested and demonstrated as part of the Fuel Performance Improvement Program includes a graphite coating applied to the inner surface of the Zircaloy cladding. The technology for producing acceptable coatings was developed starting with the selection of a suitable graphite and culminating with the coating of 200 cladding tubes for demonstration irradiations in the Big Rock Point Reactor. During the process development, methods for controlling the coating thickness and minimizing its hydrogen content were developed that were compatible with production scale operation. During the same period, the graphite coatings were characterized and their properties were determined, mainly to provide information needed for fuel design and licensing considerations. Purity, structure, coating-to-cladding compatibility, thermal conductivity, and friction/ wear characteristics were measured. Section 1 of this report describes the process development that was performed and the resulting process used to produce acceptable graphite-coated cladding. The process, though not optimized, is considered to be sufficiently defined to permit pilot-scale utilization by a fuel fabricator. Section 2 presents the results of the coating characterization work that was performed.

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1.0 GRAPHITE COATING TECHNOLOGY

1.1 INTRODUCTION

The Fuel Performance Improvement Program (FPIP) is a program sponsored by the U.S. Department of Energy (DOE) that is being performed by Consumers Power Company, Exxon Nuclear Company, and Battelle, Pacific Northwest Laboratory (PNL). The purpose of the program is to evaluate several fuel concepts that are more resistant than standard commercial reactor fuel to failure from pellet-cladding interaction (PCI) during power increases in light water reactors (LWRs). A more PCI-resistant fuel offers extended burnup potential and, hence, increased reactor operating efficiency. Early in the program, a State-of-the-Technology report⁽¹⁾ identified several design modifications with potential for improving PCI performance. Some of the modifications had already been tested under less stringent conditions⁽²⁾ and some were relatively unproven. One already-tested concept involved coating the inside surface of the Zircaloy cladding with a layer of graphite.⁽²⁾

Over a period of about two years, various technical problems associated with the application of graphite coatings were addressed and about 200 cladding tubes were coated for irradiation tests in the Halden Boiling Water Reactor (HBWR) and demonstration irradiations in the Big Rock Point Reactor (BRPR). The purpose of this report is to document the technology that was developed and suggest areas where future manufacturing development would be most fruitful if program results favor commercialization of this fuel concept for LWR application.

1.2 SELECTION OF GRAPHITE DAG

After the decision was made that a graphite dispersion would be the simplest coating method, several graphite dags^(a) were evaluated using the following criteria:

1. The dag shall not contain impurities that are objectionable from a nuclear or a compatibility standpoint.

(a) All of the graphite dags were obtained from Acheson Colloids Co., Port Huron, Michigan. The term "dag" is an acronym for deflocculated artificial graphite.

2. The dag shall wet the cladding and adhere to it sufficiently to tolerate processing and pellet loading.
3. The dag shall not contain materials that are difficult to dehydrate or that are highly hygroscopic.
4. The dag shall be commercially available.

It was difficult to find a dag that met these requirements. For instance, a dag designated as RD-5 bonded tenaciously to the cladding; however, the bonding agent was sodium silicate. Sodium silicate, being highly hygroscopic, absorbed excessive moisture after outgassing. Another dag (Aquadag) that was evaluated did not contain any added binders; hence it tended to flake and peel from the smooth inside surface of the cladding. The final choice of the first evaluation was Dag 154,^(a) an isopropanol-based dag containing a thermoplastic binder that decomposes to essentially pure carbon during vacuum outgassing. This material was, in fact, used for the first coated fuel rods that were used in the Halden Series H-1 irradiation tests. However, during pellet loading, it was noted that the adherence of this dag coating was not as good as desired. Consequently, to avoid excessive scarring of the coating, pellets were inserted in groups of five and the cladding was rotated 90° after each fourth of the pellet stack was loaded.

A search for a more durable dag identified a product that is used primarily as a coating for television picture tubes and designated by the supplier as Dag 4. This water-based dag contained alumina and silica in gel form as a binder and was significantly more adherent and resistant to abrasion during pellet loading than Dag 154. However, Dag 4 was found to be slightly more hygroscopic than Dag 154 but less than Dag RD-5. Therefore, Dag 4 was selected as the preferred coating type and was used for the remainder of the coated cladding used in the FPIP. Even though the application method used was essentially the same as for Dag 154, thickness control was more difficult and more stringent moisture control measures were required than for Dag 154. The work performed on graphite dags included some characterization which is important but not process-related. This information is presented in Section 2.

1.3 COATED CLADDING THICKNESS SPECIFICATION

The specified thickness for graphite applied to the inside surface of Zircaloy cladding was a compromise between the required gap size considering the surface roughness of surface-ground UO_2 pellets and Zircaloy cladding (as a minimum) and by thermal conduction and the need to minimize the amount of water and hydrogen contained in a fuel rod (as a maximum). Engineering judgment led to a specification of $6.4 \pm 1.3 \mu\text{m}$ average coating thickness. A maximum tolerance of $\pm 4 \mu\text{m}$ from the specified average was permitted at individual points. The specification was established before a large experience base could be accumulated and it proved difficult to meet in practice. The need for a group of demonstration fuel rods with very uniform coating thickness was met, but the thickness tolerance specification might be relaxed for commercial practice.

1.4 COATING APPLICATION PROCESS FLOWSHEET

The coating application and processing technology is illustrated by the process flow chart shown in Figure 1.1. The major steps are described separately in a narrative together with pertinent background information that describes why a particular step was performed in the manner stated.

1.4.1 Coating Application Method

Several possible methods for coating the inside surface of a cladding tube with graphite were considered. The most sophisticated of these is probably pyrolytic deposition in which a hydrocarbon gas is thermally cracked, resulting in the deposition of carbon on the surface. Temperature, concentration, and gas flow can be adjusted to produce carbon of any desired density and crystallinity, from sooty carbon to dense crystalline pyrolytic graphite. The process is complicated and Zircaloy cladding cannot be subjected to the high temperature (1200°C minimum) necessary to produce dense crystalline coatings. For these reasons, this method was not seriously considered.

A much simpler application technique uses a dispersion of fine ($\sim 5 \mu\text{m}$) graphite particles in a liquid carrier. This dispersion is known as a dag and is commonly used for applying graphite coatings to metal surfaces, primarily for use as a lubricant. Several means of applying the dag were considered;

FIGURE 1.1. Process Flow Chart for Applying Graphite Coating to the Inside Surface of Cladding

among them were spraying, swabbing, and flood coating. After considering the possibilities, it appeared that the most uniform thickness could be obtained by flood-coating, i.e., immersion of the surface in dag. Although this could be accomplished by filling and draining a tube, better process control could be obtained by moving a silicone rubber dam downward through the tube. The dam supports a predetermined (4 cm) column of dag and is moved along the inside of the tube with the plug. Figure 1.2 shows a schematic of the dam and the method used for controlling the movement of the dam down the tube.

The tube to be coated was mounted in a vertical position in a simple U-beam fixture that provided a track for the magnet. The silicone rubber dam was placed inside the tube approximately 10 cm from the top end. The dag was removed from the mixer and injected into the tube with a 10 cc hypodermic syringe. The magnet was then pulled downward at a fixed speed (~ 10 cm/sec) using an electric motor and chain drive. The dam followed the magnet, and the dag coated the inner tube wall by surface wetting. The dam was pulled from the tube bottom and the excess dag was allowed to drain and dry.

Predictable control of coating thickness proved to be the most elusive and difficult development problem. This problem was aggravated by the fact that Dag 4 is a thixotropic fluid, meaning that it is gel-like and its viscosity is dependent on shear rate. During flood-coating of cladding tubes

FIGURE 1.2. Method for Wetting the Inner Cladding Surface with Dag.

with Dag 4, the liquid dag does not dry immediately but, due to the high humidity inside the tube, continues to drain from the bottom end. Thus, the thickness of the coating obtained is highly dependent on the fluid properties of the dag. A substantial development effort was expended to determine the relationship between the solids content of the dag and the thickness of the resulting coating. Figure 1.3 shows a plot of some of the data obtained and also serves to illustrate the variability in results when slightly different application techniques were used. Other factors such as draining and drying

FIGURE 1.3. Graphite Coating Thickness as a Function of Solids Content

times were found to be more important than first realized and caused unpredictable results when the process parameters determined in the laboratory were applied to the coating of a large number of tubes and different length tubes. Initial reject rates based on thickness specifications were ~50%, which would be unacceptable for a commercial process. During subsequent coating of a large number of cladding tubes for the BRPR demonstration fuel, the following controls were implemented which significantly decreased the reject rate:

- The dag was continuously agitated to eliminate any tendency toward either settling or gelation.
- The mixing container was modified to prevent air exposure while withdrawing dag.
- Draining and drying times were carefully standardized at 10 and 5 minutes, respectively.
- At the beginning of each work day, a trial group of tubes was coated and the thickness was measured to confirm that the production would meet the specifications or indicate a need for an adjustment of solids content.
- Tubes were coated sequentially to identify trends in coating thickness with time, and the solids content of the dag was monitored to relate this parameter to coating thickness. Solids contents of 11.8, 11.9 and 11.7 wt% were used in the BRPR tube coating campaign; in good agreement with the middle line of Figure 1.3.

Although it was believed that some of these measures were not necessary, there was insufficient time to determine which factors were most important. The acceptance rate increased to >90% as a result of these measures, and the process was considered to be sufficiently well developed for satisfying the needs of FPIP. However, it is anticipated that further manufacturing development would be desirable for scaled-up production quantities.

1.4.2 Coating Drying

Drying of the dag coating using heated air was shown to be an important step. Drying should be done immediately after coating (<15 min) so that the

dag lost through drainage is minimized. A commercially available hair dryer^(a) that supplied 80°C air at 0.7 liters/sec was used. After draining in the vertical position for ten minutes, the 1.8 m-long coated tubes for BRPR were connected to a three-outlet manifold (air-flow downward) and air dried for 5 minutes. In order to ensure a constant, reproducible flow of heated air, three tubes were connected to the manifold at a time. These parameters were appropriate for the 1.8-m BRPR cladding (9.449 mm I.D), but might need adjustment for other cladding sizes.

If the coating was dried too fast, the resulting water gradient within the coating resulted in nonuniform shrinkage and crazed coatings. With the total output of the dryer (2.3 liters/sec) channeled through one tube ~1 cm ID, the coating sometimes cracked. The drying process was accomplished at close to the maximum rate possible without creating coating defects.

1.4.3 Coating Thickness Measurement

Measurement of the thickness of a graphite coating 6 μm thick was difficult. The coating was easily damaged; hence, a high-precision, noncontacting measuring device was required. These factors led to the use of air-gauging as the measurement method. In air gauging, air is passed through a number of jets in a precision plug and the impact of these air streams on the adjacent surface creates a back pressure that can be measured. Thus, the probe floats on a cushion of air in the manner of an air bearing and does not physically contact the tube. With suitable calibration and under carefully controlled conditions, very precise and accurate measurements of the inside diameter can be obtained.

The inside surface of cladding did not represent ideal measuring conditions because the cladding was not perfectly round and the diameter changed continuously along the length. Because the tubing had been run through a Pilger tube-reduction step during manufacture, an approximately sinusoidal variation in diameter occurred along the length of the tube. Accordingly, in order to obtain reproducible measurements, the maximum and minimum inside diameter was measured at each of five locations along the full length. The locations were indexed so they could be relocated with a precision of ± 0.5 mm. Intuitively, it might be expected by virtue of the coating method used, that

(a) General Electric Superblower Hair Dryer, Model No. 04-481

the variation in coating thickness with axial location in the tube would be small. This was found to be the case, and it is possible that the number of measurement sites could be decreased.

The inside diameter of the tubes was measured with an air gauge before and after coating. The procedure was to measure each tube horizontally, at five equally spaced locations. At each location the tube was rotated several times and the maximum and minimum diameters were recorded. The procedure was repeated at the same locations after coating. The maximum diameter was subtracted from the maximum for the noncoated condition and divided by two to determine coating thickness; the same was done for the minimum readings. All ten thickness measurements were averaged to obtain a tube average. The specified tube average was $6.4\text{ }\mu\text{m}$: the maximum/minimum limits of individual points were 10.4 and $2.4\text{ }\mu\text{m}$.

The air gauging equipment used^(a) was highly precise (standard deviation of max/min cladding diameters was $0.38\text{ }\mu\text{m}$, for 10 measurements) and accurate (set rings accurate to $0.8\text{ }\mu\text{m}$ were used to set up and calibrate the equipment). Measuring coating thickness is the most time consuming operation in the entire coating process. The measurements must be made with great care, and the time required is estimated to be about 80% of the labor costs of producing graphite-coated cladding. Thus, a technique that measures the coating quickly and directly would be highly desirable.

Any mechanical technique for measuring the coating thickness would involve applying a contact force to the coating, which because of the low coating density ($\sim 60\%$ TD), would possibly reduce the thickness and damage the coating. Measurement of the thickness by the beta backscatter method requires essentially no contact pressure because the interrogation is by means of beta particles. The beam of beta particles is attenuated by the number of carbon atoms in its path, which is related to the thickness and density of the coating. The density of Dag 4 has been measured at 1.55 g/cc by mercury displacement measurements made on bulk ($\sim 0.5\text{ cm}^3$) deposits, and the density of a $6\text{ }\mu\text{m}$ -thick deposit is assumed to be the same as that of the bulk deposit.

(a) Linear reading Freeland Air Comparator, 1 division = $1.3\text{ }\mu\text{m}$, Freeland Gauge Co., Detroit, MI 43227

Although the results of this work indicate a need to improve the method for measuring coating thickness, evaluation of beta backscatter as a method for measuring coating thickness was not pursued. A single U.S. manufacturer^(a) has experience in manufacturing the equipment.

1.4.4 Coating Outgassing

The purpose of outgassing the graphite coated cladding is to reduce the hydrogen contained in the coating in the form of water to an amount that is no more than half of the limit established for the fuel. Considering the relative amounts of fuel and graphite present in a fuel rod, the average water content of the graphite coating, after application of 95/95 statistical factors, is limited to $<6.5 \text{ ppm}^{(b)}$ on a fuel weight basis. In order to reduce the water concentration to an acceptable level, the coated tubes were vacuum outgassed at a pressure $<1.3 \times 10^{-3} \text{ Pa}$ ($1 \times 10^{-5} \text{ torr}$) and at the highest temperature that would not affect the mechanical properties of the cladding in 24 hours. This temperature was determined to be 425°C after experiments demonstrated that the yield strength of the cladding was not affected by a temperature/time combination less than $450^\circ\text{C}/51 \text{ hr}$. The temperature in the vacuum outgassing furnace was controlled at $425 \pm 25^\circ\text{C}$. The outgas cycle of $1.3 \times 10^{-3} \text{ Pa}/425^\circ\text{C}/24 \text{ hr}$ should not be considered as the "optimum" or "recommended" cycle because it is considered to be quite conservative. Some experiments were performed to evaluate the effect of higher or lower temperatures and higher pressures on outgassing results. The results, summarized in Table 1.1, indicate that lower temperatures and higher pressures have the

(a) UPA Technology Inc., Syossett, N.Y. 11791.

(b) ENC specifications allow 1.5 ppm hydrogen on a fuel weight basis from all sources within the rod. Half of this hydrogen is assumed to originate in the fuel, which leaves 0.75 ppm of hydrogen from the coating, neglecting all other contributions. In a BRPR fuel rod, the ratio of fuel weight/coating weight is $1140 \text{ g}/0.55 \text{ g} = 2,073$. Therefore, $2,073 \times 0.75 = 1,555 \text{ ppm}$ hydrogen is the maximum amount allowed in the coating. Hydrogen weight is $1/9$ the weight of water, so $14,000 \text{ ppm}$ of water would be the maximum permitted in the coating. This calculation ignores the contribution of other sources of hydrogen in the coating, which has been determined to be about 2 cc/g of coating, or about 179 ppm of additional hydrogen. Because water is easier to monitor and control than hydrogen, the water concentration was used as the basis for establishing the amount of hydrogen contained in the coating.

TABLE 1.1. Effect of Outgassing Parameters on Water Content of Dag 154

Outgassing Conditions			Water Content on Fuel Wt. Basis, ppm
Temperature, °C	Pressure, Pa	Time, hr	
450	1.3×10^{-3}	24	1.6
400	1.3×10^{-3}	24	3.5
450	1.3×10^{-1}	24	3.1
400	1.3×10^{-1}	24	4.8

expected effect of increasing the water content of the graphite. Further experimentation would be necessary to define the optimum outgassing cycle for quantity production. It is possible that the pressure specifications could be relaxed to ~ 3 Pa permitting the use of only a cold trap and mechanical pump; however, this was not evaluated.

A 15-cm diameter tubular vacuum furnace was used to outgas the graphite coating on the cladding tubes. The procedure involved loading about 20, 2-m long BRPR cladding tubes into the vacuum furnace. Initial water removal was done at about 100°C during pumpdown. When a pressure of about 7 Pa was reached, the diffusion pump was turned on and the heating cycle was started. The temperature was increased in 50°C increments until the required set point of 425°C was reached. In this way, the pressure did not rise above 1.3×10^{-2} Pa during the heating cycle. After the required 24 hr at temperature, the furnace was turned off and allowed to cool under vacuum.

The possible effect of the outgassing cycle on the thickness of the graphite coating was evaluated by measuring coating thickness before and after the outgassing cycle. The measurements indicate no change in the graphite film thickness due to heating. The data from these measurements are summarized in Table 1.2.

1.4.5 Welding of Coated Cladding

To eliminate the graphite coating as a potential source of weld contamination, the end weld cap regions were thoroughly scrubbed with cotton swabs moistened with distilled water (ethanol in the case of Dag 154) prior to

TABLE 1.2. Graphite Coating Thickness Before and After Outgassing

Tube No.	Average Thickness, microns						Difference
	Before Cure			After Cure			
	Max.	Min.	Avg.	Max.	Min.	Avg.	
209	5.59	5.59	5.59	5.54	5.16	5.36	-.23
153	5.79	6.60	6.20	6.05	6.43	6.25	+.05
145	5.46	6.48	5.97	5.79	6.22	6.02	+.05

outgassing. This cleaning was done after drying of the coating but before outgassing, because removal of the coating was much easier at this stage of the process.

This cleaning operation was adequate as no welding difficulties were encountered with coated cladding. The welding parameters used were the same as those used with regular noncoated cladding.

1.5 COATING MOISTURE CONTROL AND ANALYSIS

Because the Dag 4 coating tends to be hygroscopic, care must be taken to limit air exposure after the outgassing step. To accomplish this, handling procedures were implemented to assure that the inside of the coated, outgassed cladding tubes was never exposed to atmospheric air for more than a few seconds. During handling operations, tight-fitting temporary plugs (Figure 1.4) were used to seal the exposed ends of the cladding tubes, and pellet loading and welding were performed in dry air atmosphere glove boxes containing <100 ppm water. Even in the glove boxes, exposure times were minimized by use of the temporary plugs.

It was not possible to obtain a valid sample of the graphite coating for water analysis once it was applied to the tubing. To ensure that the moisture specifications were met, graphite dag ground to -40 +60 mesh was outgassed in separate shorter (46 cm) lengths of cladding and carried through all the processing steps after outgassing as a production control. The dag powder was then analyzed for water and compared with the as-outgassed graphite. Table 1.3 summarizes the results of a processing simulation, and shows that water sorption under these conditions was minimal.

FIGURE 1.4. Sectional View of Temporary End Plug

After outgassing at 425°C, the coating was known to contain about 2 cc/g hydrogen, so the usual procedure for water analysis was modified. The water analysis was performed by use of a P_2O_5 cell that is not sensitive to hydrogen. The gas released from the graphite sample during heating at 800°C was first passed through a Cu-CuO cell at 300°C to convert all the hydrogen to water, then routed to the P_2O_5 water detector. When samples were exposed to room air for more than a few seconds during transfers, their water content was above the specification limit. Consequently, all transfer operations were done in atmosphere-controlled gloveboxes containing <100 ppm H_2O .

TABLE 1.3. Water Content and Sorption of Outgassed Dag 4
Blind Tube* Exposure Results

Condition	Average Water Content ($\bar{x}$) on Fuel Wt. Basis, ppm	σ	$\bar{x} + 4.2\sigma$, ppm ^(a)
As-outgassed 425°C, 24 hr, $<1.3 \times 10^{-2}$ Pa	3.79	0.79 ^(b)	7.10
Loading box + 15 min N ₂ ^(a) @ 20 ppm H ₂ O	3.63	0.47	5.63
Loading box + 1 min. air ^(a) @ 39% RH	3.79	0.42	5.58

*Tube length: 46 cm, closed one end
Dag: -40 mesh, spread along length
All analyses in quintuplicate

(a) For statistical significance at the 95/95 level when analyses are done in quintuplicate, the standard deviation (σ) is multiplied by 4.2 and added to the average.

(b) Single deviating point responsible for high σ .

The welding operation for coated cladding involved an unavoidable air exposure of about 10 seconds during insertion of the loaded rods into the welding chamber. Concern about the effect of this exposure was alleviated because air circulation inside an open-ended fuel rod containing pellets is restricted by the pellets. The fact that limited air exposure under these conditions is of no consequence was substantiated by the results of air exposure experiments (Table 1.4) performed under various conditions to evaluate the effect of short time, room air exposure on moisture pickup of graphite samples. In these experiments, the atmosphere was much more accessible to the graphite samples than if the graphite was applied to the inside surface of a tube because the graphite samples were contained in a relatively shallow pan (diameter/depth ~1) instead of in a long tube as in Table 1.3, which accounts for the marked difference in the results. The increased tendency of Dag 4 to sorb water compared to Dag 154 is also evident from the data.

TABLE 1.4. Results of Exposure of Outgassed Graphite
Dag Samples to the Indicated Atmospheres

<u>Dag Type</u>	<u>Exposure Atmosphere</u>	<u>Time</u>	<u>Water Content Fuel Weight Basis, ppm</u>
154	As-outgassed, 425°C, 24 hr, $<1.3 \times 10^{-10}$ Pa	--	1.4
154	N ₂ glove box, 20 ppm H ₂ O	15 min	1.6
154	N ₂ glove box, 20 ppm H ₂ O	1 hr	2.2
154	N ₂ glove box, 20 ppm H ₂ O	24 hr	3.0
4	N ₂ glove box, 20 ppm H ₂ O	15 min	6.0
4	N ₂ glove box, 20 ppm H ₂ O	1 hr	4.1
4	N ₂ glove box, 20 ppm H ₂ O	24 hr	9.1
4	He glove box, 50 ppm H ₂ O	15 min	8.1
4	Air, 52% relative humidity	1 min	8.5
4	Air, 52% relative humidity	5 min	9.2

1.6 CONCLUSIONS AND RECOMMENDATIONS

- By the methods discussed in this report, Zircaloy cladding can be coated with graphite as a reproducible, thin, and uniform layer.
- Though the process is sufficiently developed for the application of graphite coating to a significant number of cladding tubes, i.e., on the order of a few hundred, the process has not been fully optimized. There are probably several areas of the process in which improvements could be made or unnecessary operations eliminated without seriously degrading the quality of the coating. Among these are an improved method for measuring the coating thickness and optimization of the outgassing cycle.

TABLE 1. Results of Exposure of Uncoated Granite to Various Agents

Run Type	Exposure Agent	Time, days	Water Content, %
104	As-outgassed, 45°C, 24 hr, 10^{-10} g	—	1.4
104	H ₂ glove box, 20 ppm H ₂ O	15 min	1.5
104	H ₂ glove box, 20 ppm H ₂ O	1 hr	2.2
104	H ₂ glove box, 20 ppm H ₂ O	24 hr	2.0
4	H ₂ glove box, 20 ppm H ₂ O	15 min	0.0
4	H ₂ glove box, 20 ppm H ₂ O	1 hr	4.1
4	H ₂ glove box, 20 ppm H ₂ O	24 hr	0.1
4	H ₂ glove box, 20 ppm H ₂ O	15 min	0.1
4	Air, 52% relative humidity	1 min	0.5
4	Air, 52% relative humidity	2 min	0.5

CONCLUSIONS AND RECOMMENDATIONS

- By the method discussed in this report, epoxy cladding can be coated with granite as a representative, thin, and uniform layer.
- Though the process is sufficiently developed for the application of granite coating to a significant number of cladding tubes, i.e., on the order of a few hundred, the process has not been fully optimized. There are probably several areas of the process in which improvements could be made or unnecessary operations eliminated without seriously degrading the quality of the coating. Among these are an improved method for measuring the coating thickness and optimization of the outgassing cycle.

2.0 RELEVANT PROPERTIES OF GRAPHITE COATINGS

2.1 INTRODUCTION

Laboratory investigations were performed to characterize the graphite coatings and determine properties that are most relevant to fuel design and licensing needs. These investigations included determining the thermal conductivity of the coatings, characterizing their microstructure, evaluating coating purity, examining the compatibility between the graphite coating and Zircaloy, and friction and wear testing. The results of these investigations are described in Section 2.

2.2 THERMAL CONDUCTIVITY OF GRAPHITE COATINGS

The presence of a thin layer of graphite on the inner surface of the cladding represents an additional thermal barrier to the fuel. Hence, the thermal conductivity of the layer was determined in order to quantify the effect. The technique used was the flash method for measuring thermal diffusivity;⁽³⁾ the resulting diffusivity data were converted to thermal conductivity (K , $W/cm^{\circ}C$) by use of the relation $K = 4.86 \alpha \rho C_p$ where α = thermal diffusivity (cm^2/sec), ρ = density (g/cc), and C_p = heat capacity ($cal/g^{\circ}C$).

Diffusivity measurements required samples that were 0.3 to 0.6 mm thick. This was difficult to achieve because Dag 154 deposits of this thickness curled and fractured. To avoid this problem, the deposit was formed by sequential dipping of a removable (paper) substrate; about 100 dippings were required to achieve the necessary specimen thickness. The Dag 4 specimen was prepared as a one-step casting that was dried slowly to minimize the amount of warpage. Thermal diffusivity measurements were made with no further outgassing treatment of the specimens. However, specimens were outgassed in the diffusivity apparatus itself by heating to $500^{\circ}C$ in high-purity argon. Initial heatup data were ignored because of outgassing effects and no changes were measured in thermal diffusivity upon further cycling after the initial heatup.

The thermal diffusivity values were converted to thermal conductivities by using the reported C_p for graphite⁽⁴⁾ and the measured density of 0.95 g/cc for outgassed Dag 154 and 1.55 g/cc for outgassed Dag 4. Density measurements

were made using the mercury displacement method. The thermal conductivity results plotted in Figure 2.1 show that the thermal conductivity of Dag 4 is a factor of five higher than the conductivity of Dag 154. This difference is attributed to the difference in the structure of the as-deposited coatings. The graphite particles in Dag 4 tend to be larger and more equiaxed and are oriented more randomly than those of Dag 154, which tend to lie parallel to the cladding surface. The suspected effect of particle orientation on thermal conductivity of the coatings is illustrated in Figure 2.2.

FIGURE 2.1. Thermal Conductivity of Dags 4 and 154 as a Function of Temperature.

FIGURE 2.2. Schematic Diagram of Dag Deposits. Differences in particle orientation may explain the reason for the higher thermal conductivity of Dag 4 compared with Dag 154.

The effect of the graphite coating on the overall resistance to heat transfer from a fuel rod is minor. Calculations using the measured values of thermal conductivity show that at a linear heat generation rate of 32.8 kW/m (10 kW/ft), the maximum expected ΔT across a 6.4 μm thick graphite coating is about 20°C, even using the lower conductivity value for Dag 154.

The above calculation applies to beginning-of-life conditions only. In the case of a fuel rod that has been irradiated to a significant burnup level and thus contains cracked pellets that are in hard contact with the coated cladding, the graphite coating may enhance heat transfer by bridging gaps caused by asperities existing between the fuel and cladding. The magnitude of this effect was not evaluated.

2.3 MICROSTRUCTURE OF DAG COATINGS

Microscopy was performed to characterize the two types of dag coatings and to note their differences. Conventional sample preparation techniques utilized for optical metallography were not satisfactory for examining the

graphite layer when it was present at normal thicknesses because the layer was destroyed during polishing. This problem is usually remedied by vacuum-impregnation with a low-viscosity resin,^(a) followed by using otherwise normal sample preparation techniques. However, impregnation caused the graphite coating to swell to about three times its normal thickness because of resin inclusions in the graphite porosity.

Examination of the coatings by SEM was much more satisfactory. When viewed from the top surface, the graphite particles in Dag 4 appeared larger and more equiaxed than the Dag 154 (Figures 2.3 and 2.4). By examining fractured coating sections on-edge with the SEM (Figure 2.5), it was possible to compare direct thickness measurements with air-gauge measurements. The results of SEM measurements confirmed air-gauge measurements taken at the same location; i.e., the average of the SEM measurement was 7.2 μm versus an average air-gauge reading of 6.4 μm at the same site.

SEM examination of the graphite particles after elimination of most of the binder by dilution shows that the Dag 4 particles (Figure 2.6) are larger and more equiaxed than the Dag 154 particles (Figure 2.7) and that they lie in a narrower size range. Dag 154 particles are more plate-like in character, which probably explains why they tend to lie more parallel to the cladding surface.

The binder used in Dag 154 is converted to carbon during outgassing. The binder used in Dag 4 is composed of Al_2O_3 and SiO_2 in a gel form and is easily detectable using the SEM. Due to the difference in electron production efficiencies between aluminum and silicon, the alumina is highlighted in SEM micrographs (Figure 2.4); a randomly chosen area reveals more Si than Al. The binder is intimately mixed in the dag so that random location analysis of Dag 4 always shows Al and Si. In order to examine the binder separately, a sample of Dag 4 was oxidized 70 hr at 700°C to eliminate the graphite, which resulted in a loss of 66.5% of the original dry weight. Apparently, the oxide binder residue (Figure 2.8), is sufficiently continuous that it forms a coherent skeletal structure.

(a) One such resin is Maraglas 555, Acme Chemical and Insulation, New Haven, CT.

FIGURE 2.3 As-Deposited Top Surface of Dag 154.

FIGURE 2.4. As-Deposited Top Surface of Dag 4. The bright area contains excess aluminum.

FIGURE 2.5. Edge-on View of Dag 154 Graphite Deposit.

FIGURE 2.6. Individual Dag 4 Platelets

FIGURE 2.7. Individual Dag 154 Platelets

FIGURE 2.8. Residue from Oxidized Dag 4 Deposit (Fracture Surface).

2.4 PURITY OF DAG COATINGS

The two graphite dags evaluated for coating cladding tubes are commonly used for lubrication or for electrical conduction applications. Neither of these uses would require a very high-purity product, and they could contain impurities that are objectionable for a nuclear application. Metal impurity concentration in the dags was determined by emission spectroscopy and the concentration of halides was determined by bomb combustion followed by amperometric analysis. The results are summarized in Table 2.1. Considering that a full-length (1.8 m) BRPR cladding tube would contain only ~0.5 g of dag, no impurities are present in objectionable quantities. Alumina and silica, which amount to 33.5% of the dry weight, are present in Dag 4 as a binder. Neither of these compounds is expected to react with the Zircaloy cladding under normal temperature conditions, as described in Section 2.5.

TABLE 2.1. Analysis of Graphite Dags^(a)

<u>Element</u>	<u>Dag 154</u>	<u>Dag 4</u>
Fe	0.02	0.6
Si	0.001	10.7
Al	0.0005	4.8
Ca	0.01	1.3
Na	0.01	2.1
F	9 ppm	50 ppm
Cl	62 ppm	720 ppm
B	N.D.	N.D.

(a) All analytical results are in weight percent of the solid portion of the dag unless otherwise stated.

2.5 COMPATIBILITY OF GRAPHITE COATINGS WITH ZIRCALOY CLADDING

Graphite coating materials would not be expected to diffuse significantly into the Zircaloy cladding at the temperatures encountered during normal irradiation conditions. However, because of the extended time - 30,000 hr for four complete cycles in BRPR at $\sim 350^\circ\text{C}$ - it was considered important to evaluate the compatibility of the graphite coating and the Zircaloy cladding. Available literature on carbon diffusion into zirconium and Zircaloy^(5,6) was evaluated. Extrapolation of the relationship given by the diffusion equation $D = D_0 e^{-Q/RT}$, where both D_0 and Q have been determined experimentally for the temperature range listed, gives the diffusion coefficients summarized in Table 2.2.

By performing a standard Fick's law calculation, these values result in the diffusion profiles shown in Figure 2.9. The values of C^{14} and C diffusion into α -zirconium are from different sources and are in excellent agreement. According to Reference 6, the slower diffusion in the Zircaloy may be due to the stiffening of the zirconium lattice by the alloying elements. The results show that after 30,000 hours, the penetration depth of carbon in Zircaloy to a concentration of ~ 100 ppm, is expected to be about $15\ \mu\text{m}$. Carbon solubility (substitutional) in zirconium and its alloys is small because of the formation of ZrC (interstitial). Because carbon reduces the corrosion resistance of Zircaloy in hot acidic water, specifications for reactor-quality Zircaloy tubing call for a maximum of 100 ppm C . The presence of small amounts of carbon does not greatly influence the strength of zirconium and its alloys^(7,8) and is not likely to cause a problem in this case.

TABLE 2.2. Calculated Diffusion Coefficients

Ref. No.	Temperature Range, $^\circ\text{C}$	Diffusing Element	Solute	$D_0, \text{cm}^2/\text{sec}$	$Q, \text{cal/mole}$	$D, \text{cm}^2/\text{sec}$ (extrapolated to 400°C)
5	600-800	C^{14}	α -zirconium	3.5×10^{-5}	30,700	3.8×10^{-15}
6	600-850	C^{14}	α -zirconium	2×10^{-3}	36,200	3.5×10^{-15}
6	600-770	C^{14}	Zircaloy-2	1.4×10^{-3}	38,000	6.5×10^{-16}

FIGURE 2.9. Calculated Diffusion of Carbon into Zirconium and Zircaloy-2 after 30,000 hr at 400°C.

In addition to the consideration of possible carbon diffusion, Dag 4 contains significant amounts of alumina and silica that could possibly react with the Zircaloy. Thermodynamically, zirconium could reduce alumina at very high temperatures (above $\sim 1300^\circ\text{C}$) and it could reduce silica at any temperature. The possibility of this reaction occurring at temperatures of interest ($\sim 400^\circ\text{C}$) and at LOCA-type temperatures ($\sim 1200^\circ\text{C}$) was examined experimentally. Test capsules were heated to 485°C to simulate the effects of a normal 30,000-hr exposure in a laboratory experiment of less than 100 hr duration. Results are summarized in Table 2.3. A pair of optical micrographs of control and graphite-coated specimens after heating to $600^\circ\text{C}/37$ hr are shown in Figure 2.10. No reaction zone is visible, and no aluminum or silicon was present in the metal as indicated by a thorough SEM-microprobe examination of the surface of a specimen exposed at $1200^\circ\text{C}/4$ sec. Apparently, the kinetics are not sufficiently rapid to cause a significant reaction.

TABLE 2.3. Conditions and Results of Dag 4/Zircaloy Compatibility Experiments

Contents	Temperature/Time	External Appearance	Metallographic Results	SEM Results
Empty (control)	600°C/37 hr	No change	No reaction	--
Coated, UO ₂ pellet	600°C/37 hr	No change	--	No Al or Si in the area examined
Non-coated, UO ₂ pellet	1200°C/4 sec	Capsule intact but bulged from pressure, large grains	No reaction layer (not sectioned adjacent to UO ₂)	--
Coated, UO ₂ pellet	1200°C/4 sec	Capsule intact but bulged from pressure, large grains	No reaction layer (not sectioned adjacent to UO ₂)	No Al or Si in the coating/cladding interface area examined
Coated, UO ₂ pellet, 10X normal amount of Dag 4	1200°C/4 sec	Capsule intact but bulged from pressure, large grains	No reaction layer (not sectioned adjacent to UO ₂)	--
Empty (control)	485°C/100 hr	No change	No reaction	--
Coated	485°C/100 hr	No change	No reaction	--
Coated, 10X normal amount of Dag 4	485°C/100 hr	No change	No reaction	--

2.6 FRICTION AND WEAR TESTING

Because pellet-to-cladding friction may contribute to the effectiveness of graphite coatings to improve PCI behavior, a series of tests was performed to evaluate friction and wear characteristics of the graphite coatings. Dag 154 was evaluated and used as the preferred coating for the inside surface of the cladding in the first series of irradiations being conducted in the HBWR. Subsequently, after further evaluation of potential graphite coating types, Dag 4 was substituted for Dag 154 and was selected as the preferred coating because of its superior adherence characteristics and abrasion resistance

ZIRCALOY
CLADDING

20 μm

(a)

ZIRCALOY
CLADDING

20 μm

(b)

FIGURE 2.10. Photomicrograph of Control (a) and Dag 4-Coated Zircaloy Cladding (b) Samples after Heating to 600°C/37 Hr. No reaction is observable.

during pellet loading. For comparison purposes, both Dag 154 and Dag 4 were included in the friction and wear evaluations.

A pin-on-disc type of apparatus, utilizing a round-tipped (1.3 cm radius) UO_2 pellet riding on coated or noncoated Zircaloy discs, was used for these tests. With this apparatus, a variable load was applied and measurements were made over sliding distances that were long and speeds that were high compared to the differential pellet-cladding movement that actually occurs in a fuel rod during irradiation. Static friction, i.e., the friction force just before movement occurs between the UO_2 pellet and the Zircaloy disc, was also measured.

It was recognized that the test conditions that would most closely simulate those that exist between UO_2 pellets and Zircaloy cladding in an actual fuel rod are high load and small differential movement at very low speeds. However, it was expected that the test results obtained over long sliding distances would: (1) provide a relative measure of the effectiveness of the different graphite coatings to reduce the pellet-to-cladding friction and (2) provide a relative comparison of the friction and wear behavior of coated and noncoated cladding. The static friction results are considered to be more representative of the physical situation that exists between the fuel and cladding in an actual fuel rod.

During the performance of each test, a coating was arbitrarily assumed to have failed when the friction coefficient reached a value of 0.2. This failure criterion was verified as being reasonable by inspecting the disc wear track after testing. With no coating failure during the test, the wear track was narrow with only smooth shiny graphite visible. At failure, the coating was penetrated and the wear track clearly showed bare Zircaloy. Further testing after failure produced a wide wear track of bare Zircaloy metal with UO_2 embedded in the surface, and the rider showed a correspondingly large wear scar. Coating performance was also evaluated by calculating the amount of UO_2 worn from the rider during the test, as indicated by the diameter of the wear scar on the pellet tip.

The friction and wear test results for conditions that are most representative of those existing within an operating fuel rod, i.e., dry helium at about 300°C, are summarized in Table 2.4. To obtain wear data, testing was performed in accordance with a standard sequence of 543 revolutions of the disc regardless of the frictional force produced. The results indicate:

- Both types of graphite coating reduce the friction coefficient compared with a noncoated surface,
- The wear characteristics of both graphite coatings are similar under these test conditions, and
- UO_2 rider wear is increased when a graphite coating is present.

The first result is expected, and the third is consistent with the fact that graphite can become an abrasive under dry, high-temperature conditions.

The mechanical loading conditions that exist between the fuel pellet and the cladding during PCI are such that static rather than kinetic friction behavior is probably most important. Consequently, the test apparatus was modified so that the tractive force could be increased slowly until displacement occurred. The static friction coefficient was calculated using the peak force recorded before pellet sliding occurred; results are summarized in Table 2.5. In the case of Dag 4, no peak load was discernible; thus the reported coefficients are kinetic.

Given the extremely dry atmosphere and high pellet-to-cladding loading conditions that exist within an operating fuel rod, it is doubtful that either dag would reduce the pellet-to-cladding friction coefficient significantly over long sliding distances. However, the very small differential fuel-to-cladding movements that occur in a fuel rod would permit graphite to function as a lubricant or a parting layer to prevent hard pellet-cladding engagement or bonding. Hence, it is indicated that the graphite coating applied to the inside surface of the cladding reduces pellet-to-cladding friction enough to allow a small amount of slippage. This slippage would tend to distribute localized cladding stresses and thus reduce the mechanical contribution to PCI-type failures.

TABLE 2.4. Friction and Wear Test Results for a Standard Sequence of 543 Revolutions

Test No.	Zircaloy Disk Coating	Test Conditions	Life of Coating in Revolutions	Friction Coefficient, μ	Wear of UO ₂ Rider in g Worn per m of Sliding Distance (1 rev = 6.3 cm)
15	None	300°C, helium ^(b)	0	0.19-0.34 ^(c)	2.1×10^{-6}
16	None	300°C, helium	0	0.21-0.32	4.2×10^{-6}
17	None	300°C, helium	0	0.21-0.42	7.0×10^{-6}
18	Dag 154 ^(a)	300°C, helium	3.8	0.5-0.42	9.5×10^{-6}
19	Dag 154	300°C, helium	17.5	0.03-0.24	9.9×10^{-6}
20	Dag 154	300°C, helium	17.5	0.01-0.31	7.5×10^{-6}
21	Dag 4	300°C, helium	8.5	0.02-0.37	7.0×10^{-6}
22	Dag 4	300°C, helium	17.5	0.03-0.42	10.0×10^{-6}

- (a) Coatings were ~6 μ m thick and outgassed for 24 hr at 425°C prior to testing and were applied to a Zircaloy surface ground through 600 grit.
 (b) Helium was backfilled through an 825°C zirconium getter into an evacuated chamber.
 (c) The coefficients of friction listed in the left hand column are values obtained at the beginning of the test, whereas the values listed in the right hand column were obtained at the end of the test.

TABLE 2.5. Static Friction Results

Coating and Testing Conditions	Static and Kinetic Friction Coefficients (μ_s and μ_k) from Static Friction Test)	
	3.3- to 13.2-kg load	26.3-kg load
Noncoated, 300°C helium	$0.39_s - 0.31_k$	$0.32_s - 0.28_k$
Dag 154, 300°C helium	$0.06_s - 0.05_k$	0.035_k
Dag 4, 300°C helium	0.06_k	0.06_k

s = static
 k = kinetic

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