NUCLEAR GRAPHITE DEVELOPMENT, OPERATIONAL PROBLEMS, AND RESOLUTION OF THESE PROBLEMS AT THE HANFORD PRODUCTION REACTORS[a]

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Abstract

The first reactors designed for the whole-scale production of special nuclear materials were the graphite-moderated B, D, and F Reactors at the Hanford Engineering Works, northwest of Richland, WA (USA). These three Hanford Production Reactors (and the three generations of horizontally-tubed, water-cooled, graphite-moderated reactors that followed them) served as test beds for the development of improved nuclear-grade graphites. They also presented challenges to reactor operations staff and the scientific-support staff, as the effects of neutron radiation manifested itself through the changes in dimensions and physical properties of the graphite moderator bars.

In-depth studies at other research establishments have greatly expanded current knowledge regarding radiation-induced changes in the chemical and physical properties of graphites, the effects of manufacturing variables (especially precursor materials) on the magnitude of the changes, and the importance of reactor design and operational variables that can minimize the impact of these changes on reactor operations. However, at the Hanford Production Reactors, knowledge regarding the effects of neutron radiation on graphite was gained by observation of the effects of those changes on the operational characteristics of the reactors, the physical distortions of the graphite moderator blocks, and the physical properties and chemical reactivities of small samples of graphite that had been exposed to neutron radiation within the reactors themselves. Each generation of production reactors was constructed (primarily) from a new grade of graphite, and was designed to operate under different conditions than was the previous generation; each generation disclosed new radiation-effects phenomena that complicated continued safe operation of the reactors, and presented new challenges to the operations and scientific-support staff at Hanford.

This paper chronicles the history of the Hanford Production Reactors, from the initial design considerations for B, D, and F Reactors through the selection of the agreed method for safe disposal of the decommissioned reactors. The operational problems that challenged the operations and support staff of each new generation of production reactors, the engineering actions and operational changes that alleviated or resolved the immediate problems, the changes in reactor design and design-bases for the next generation of production reactors, and the changes in manufacturing variables that resulted in new "improved" grades of nuclear graphites for use in the moderators of the Hanford Production Reactors are reviewed in the context of the existing knowledge-base and the mission-driven priorities of the time.

Introduction

Much has been written on the events preceding the first demonstration of a controlled, self-sustaining nuclear chain reaction in the first Chicago Pile (CP-1) on 2 December 1942, and of the subsequent scientific and engineering feats accomplished under the Manhattan Project.[b] It is sufficient, for purpose of this review, to note that the first discharge of irradiated uranium fuel from the Hanford B Reactor occurred less than two years after that historic demonstration in Chicago. Design and construction of the first three production reactors, and the massive chemical complex required to support them, was perceived as an urgent race against time where every effort must be made to ensure that everything worked the first time.

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[b] See, for example: Fermi 1952, Wigner 1946, Gerber 1993.

To ensure that everything would work the first time, several decisions were made in the early days of the Manhattan Project; among these were the decisions to utilize proven technology to the maximum extent possible, to provide back-up solutions for hypothetical problems, and to over-design the reactors. Thus, the production reactors were graphite-moderated and water-cooled, manufacturing processes for the first nuclear-grade graphites closely followed established commercial practices, an excess number of fuel channels were provided, and additional control rods were added to the three deemed necessary.

On 1 January 1947, oversight of the B, D, and F Production Reactors was transferred from the U. S. Army Corps of Engineers to the newly formed U. S. Atomic Energy Commission (AEC); in subsequent years, six additional graphite-moderated production reactors were constructed at Hanford, during three periods of time. Table 1 summarizes the startup dates and information on the specific power generation rates of the Hanford production reactors. It is especially noteworthy that the increases in "design power" for H and C Reactors was achieved without major modifications from the design used for the B, D, and F Reactors, and that the B, D, and F Reactors eventually operated with specific power levels that were more than eight-times their original design power.

TABLE 1. The Four Generations of Production Reactors at Hanford

Hanford Reactors	Startup Dates	Design Power MW	Volume of Core m^3	Specific Power, MW/m^3	
				Design	Maximum
B, D, F	9/44 to 2/45	250	654	0.38	3.38
DR, H, C	10/49 to 11/52	250, 400, 650	654	0.38, 0.61, 0.99	3.82
KW, KE	1/55 and 4/55	1850	843	2.19	5.22
N	6/64	4000	495	8.09	8.09

Table 2 shows the primary grades of graphites which were used in the central region of the cores of the various Hanford Reactors, and summarizes some of the important differences between the various grades. All of the graphites are extruded, with petroleum-coke filler and coal-tar pitch binder. Each grade was produced during a very limited period of time, using a specific source for the coke and the pitch. In general, material with the highest nuclear purity was selected for use in the central-most region (green zone) of the reactor core, slightly lower purity material was used in the surrounding region (white zone), with the less pure material being used in the fringe or reflector regions (or reserved for non-nuclear applications).

TABLE 2. Nuclear Graphites in the Cores of the Hanford Reactors

Hanford Reactors	Primary Grade of Graphite	Morphology of Petroleum Coke	Graphitization Temperature, °C	Thermal Expansion Ratio (25-425°C)
B, D, F	KC	Semi-needle	2800	
DR	KCF	Semi-needle	2800	
H	CSF	Conventional	2800	2.1
C	CSGBF	Conventional	2450	1.8
KE, KW	TSGBF	Semi-isotropic	2450	1.5
N	TSX	Needle	3000	4.0

Because nuclear purity of the graphite was of prime concern, Kendall coke and Chicago pitch were selected (primarily on the basis of their low boron content) as the preferred precursor materials to be used in the production of graphite for the first three production reactors. However, both materials were in limited supply, and Cleves coke and Standard pitch were chosen as alternative starting materials. The finished graphites were then segregated on the basis of chemical or nuclear tests.

B Reactor began operation on 26 September 1944, with only 901 of the 2004 tubes charged with uranium. Shortly after the reactor power was raised to 9 MW, it became apparent that reactivity of the reactor was decreasing; after 18 hours of operation, the power level of 9 MW could not be sustained, and the reactor was shutdown. Subsequent experiments showed that the problem was due to the buildup of xenon-135, and that continuous operation was limited to not more than about 3 MW with 901 tubes loaded. With 1500 tubes loaded (this was the design number of loaded tubes for operation at 250 MW), continuous operation was limited to 94 MW. Only 216 MW was possible with all 2004 tubes loaded with the design number of fuel elements. It also became apparent that more than three control rods were required to prevent power oscillations caused by time-dependent changes in xenon concentrations. Operations resumed with 2004 tubes loaded; but, B Reactor was not able to operate at design power until February 1945, when sufficient impurities (mostly boron) in the graphite had been transmuted to isotopes with lower absorption cross sections.

Because the graphite bars in the central regions of D and F Reactors had higher average concentrations of boron[c] than did those in B Reactor, the number of fuel elements per tube was increased; however, neither reactor could attain design power until they had been operated for some time.

By latter part of 1945, it was apparent that the center of the top shields at B and D Reactors were being pushed upwards; it was suspected that the bulging was caused by irradiation-induced growth of the central graphite.

Figure 1 shows a cross-sectional view of the components of the B, D, and F Reactor cores; the corners of all blocks were beveled to leave a passage for water from tube leaks, etc. Graphite near the tubes was at low temperatures, and was subjected to intense bombardment by high-energy neutrons. Figure 2 shows the effect of neutron radiation on KC, CSF, and TSGBF graphites at a nominal irradiation temperature of 30°C.

The graphite in the green zone of B and D Reactors was exclusively grade KC (Kendall coke, Chicago pitch) ; most of the white zone at B was grade KS (Kendall coke, Standard pitch), while that at D was grade CS (Cleves coke, Standard pitch); the expansion rate of CS graphite should be about the same as that of CSF, because it is made from the same coke and graphitized at the same temperatures. The rapid expansion of the graphite near the tubes caused the tube blocks to expand in the transverse

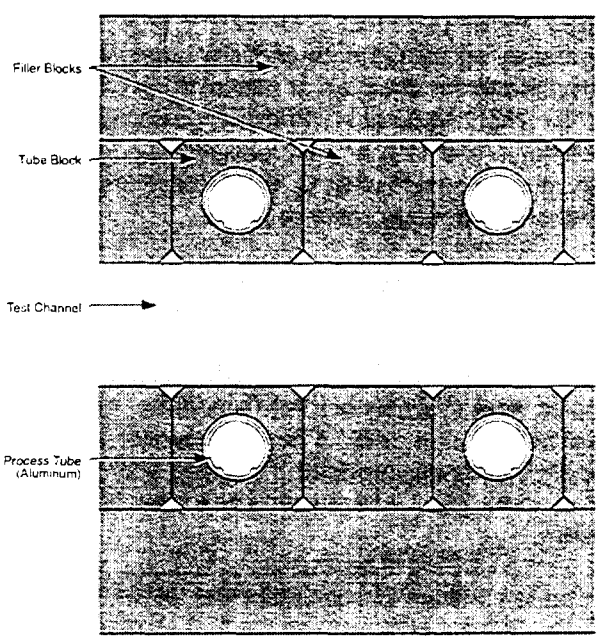

Figure 1. Cross Section of B, D, and F Reactor Cores.

c) Lewis, E. V. Pile Testing of Graphite, HW-3-1344

direction, and lift the filler layers away from the filler blocks in the tube layers. Thus, it is of no surprise that bulging of shields occurred most rapidly at B Reactor, and least rapidly at F Reactor, where all of the tube blocks in the green zone were of CS graphite.

By mid-March 1946, the top shield at B Reactor had been pushed 1.7 inches upward, and the side shields had been displaced by a total of 2 inches. At that time, B Reactor was placed on standby status (shutdown), and F Reactor was operating at reduced power levels to prolong its life. In October 1947, the AEC was given approval to build three replacement reactors[d] and two new production reactors, and permission was given to begin increasing the power levels of D and F Reactors.

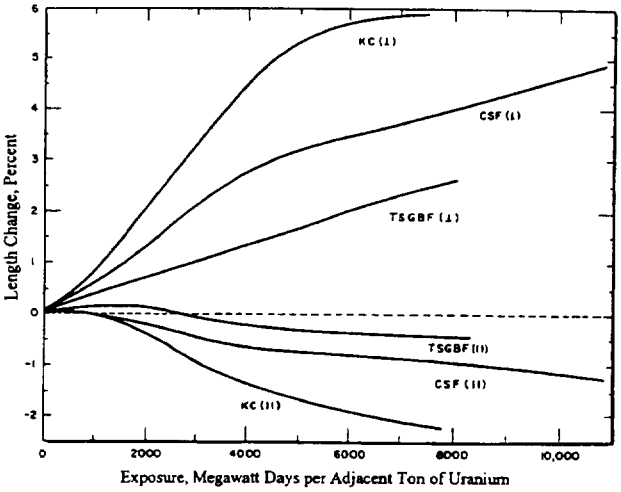

Figure 2. Length Changes of Various Graphites During Irradiation at 30°C

In January 1948, carbon dioxide (CO_2) was added to the (helium) blanket gas at D Reactor; the intent was to oxidize displaced carbon atoms and, thereby, reduce the rate of expansion. The rate of expansion was reduced; but, the reason for the reduction was that the change in gas composition increased the graphite temperatures. Increasing amounts of CO_2 were added to the blanket gas at both reactors, and to the blanket gas at B Reactor when it was restarted in July 1948.

Figure 3 shows the effect of increasing irradiation temperature on the radiation-induced expansion (transverse to the direction of extrusion) of CSF graphite (Nightingale et al. 1958). By using high concentrations of CO_2 and increasing the power levels, the expansion of B, D, and F Reactors was halted, and eventually reversed.

Stored energy had been of concern, and remained a subject of concern for more than a decade.[e] To monitor changes in stored energy and changes in physical properties (such as thermal conductivity), samples were routinely cut from moderator bars by use of hollow, core drills.[f] Rising moderator temperatures finally reduced the stored energy content to the point where it was no longer of concern.

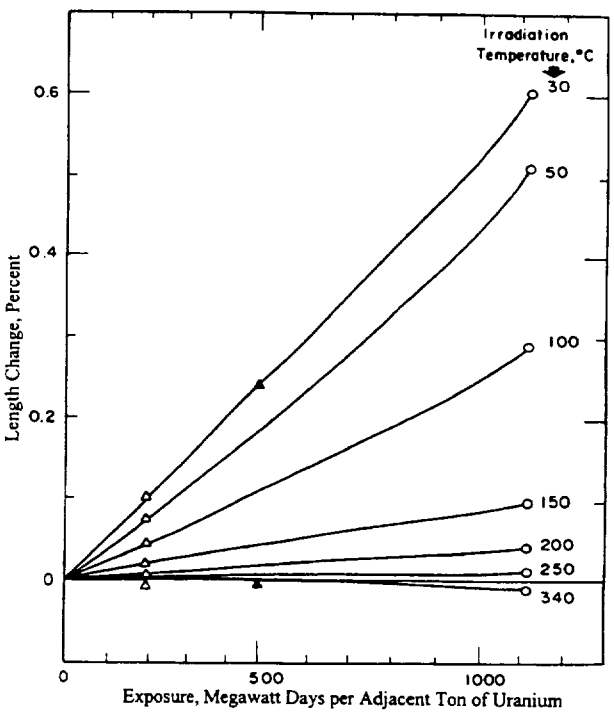

Figure 3. Length Changes of CSF (Transverse) at Several Irradiation Temperatures.

In order to continue increasing power levels, it became necessary to replace the fuel elements in the outer tube rows with lithium-aluminum alloy elements (later replaced by thoria elements); these

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d) Only DR (D Replacement) Reactor was built.

e) Morgan, W. C. 1961. Analysis of the Stored Energy Distribution in D Reactor, HW-68763 Rev.

f) Cole, J. H. 1950. Production Test 105-389-P, Graphite Boring From Process Channels, HW-19177.

elements served as a poison blanket to reduce heat-generation in the biological shields. Introduction of the poison-blankets also allowed the charging of enriched fuel elements inside the blankets, thereby flattening the side-to-side power generation rates, and reducing distortion near the moderator edges. Over-boring (reaming to increase channel diameter) of tube blocks, to flatten temperature distributions, and broaching tube blocks near the inlet and outlet, were also used to alleviate some of the problems caused by dimensional changes of the graphite.

The Second Generation of Hanford Reactors

When production of graphite for DR Reactor began, graphite purity was still considered the primary criterion for selecting precursor materials; moreover, the effects of coke morphology on dimensional-change rates (as seen in Figure 2) were not yet known. Consequently, Kendall coke and Chicago pitch were again chosen as the preferred precursor materials, with Cleves coke and Standard pitch considered as acceptable substitutes for the increasingly scarce primary precursors. During the latter part of 1947, tests of a gas-purification process had proven successful, and agreement was reached to convert some of the graphitization furnaces to gas-purification furnaces.[g] The first gas-purification furnace went into operation in January 1948; and "F" was added to the end of the grade designation (e.g., KCF) to indicate a "finished" graphite. Most of the remaining production of graphite for DR Reactor (Redding 1949a), and almost all of the graphite used in the core region of H Reactor was subject to gas purification after graphitization.

As construction of DR Reactor neared completion, it became clear that D Reactor could continue to operate, and that DR Reactor would not be able to use the cooling-water supply from D. Therefore, operation of DR was postponed to October 1950, when construction of a plant to supply cooling-water to it was completed.

The production of Kendall coke ceased about the same time as the production of graphite for H Reactor started; therefore, most of the graphite in the fueled regions of H Reactor is grade CSF (Redding 1949b); however, late in the production of graphite for H Reactor some of gas-baked carbon bars of CS material were subjected to gas purification without the intermediate graphitization step. The resultant material proved to be as pure as CSF. This change in the manufacturing process was immediately accepted, because it decreased costs and increased production rate; the resultant graphite was given the grade designation of GBF. Grade GBF graphite (later redesignated grade CSGBF) was the only grade of graphite used for the core region of C Reactor.[h]

Minor changes were made in design of the tube blocks, for each of the second-generation reactors, to provide room for free expansion of the tube blocks, so that they would not contribute to the expansion of the graphite moderator. Figure 4 shows a cross-sectional view of the components of the cores of the K Reactors, which were the third generation of production reactors at Hanford; but, C Reactor has essentially the same spacing between the tube blocks and the filler blocks as that of the K Reactors. Zirconium tubes were not, however, available when C Reactor was constructed.

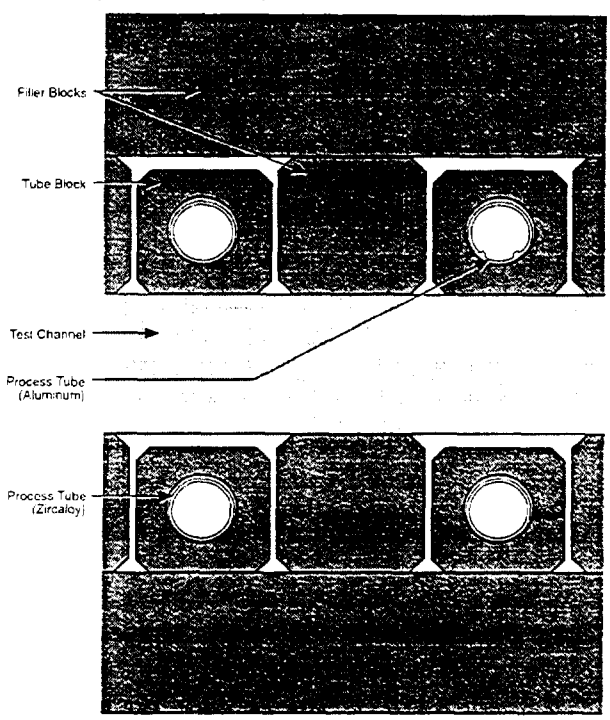

Figure 4. Cross Sectional View of the Cores of the KW and KE Reactors.

g) West, J. M. 1949. Purification of Graphite, HW-12780.

h) Boyrie, E. A., and H. A. Fowler. 1953. Report on Graphite Testing for C Pile, HW-27429.

H, DR, and C Reactors all started operation with a blanket gas of 100-percent CO_2; this gas composition, combined with the changes in design of the tube blocks, and the decision (in late 1950) to operate all reactors at the highest power attainable within approved operating limitations, raised graphite temperatures to the point that radiation-induced graphite expansion ceased to be a problem in the central regions of the Hanford Reactors (DeNeal 1970). However, it brought with it a new problem; by June 1952, the top center of H Reactor had subsided to below its pre-operational height. Much later, controlled-temperature irradiations, in test reactors having high neutron-flux densities (Nightingale et al. 1958, Helm and Davidson 1964), showed that as irradiation temperatures are increased above about 300°C the graphite contracts in the transverse direction. Figure 5 shows data on the length changes of CSF graphite during high-temperature irradiations (Cox and Helm 1969).

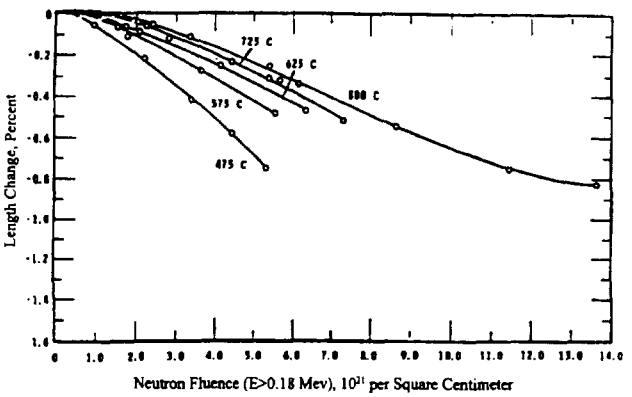

Figure 5. Transverse Length Change of CSF Graphite at High Temperatures.

One fact that has not yet been adequately explained is that the dimensions of large bars of several grades of nuclear graphite, irradiated in the Hanford Reactors, contracted at a more rapid rate than did small samples cut from bars of the same graphite grades, and irradiated under the same conditions (Nightingale and Woodruff 1964). Moreover, the filler bars in the reactor moderators contracted at the same rates as those measured on the large bars.[i]

Low temperatures, and low neutron-flux densities near the inlet and outlet faces of the reactors still resulted in expansion of graphite near the ends of the fueled region, while the central regions contracted. Thus, the tubes began to assume an "S" shaped configuration in the vertical plane, and it became increasingly difficult to charge and discharge the fuel elements. This situation was somewhat alleviated at C Reactor, where the bore of the process-tube blocks was increased near the front and rear faces, and where the introduction of short blocks with reduced bore diameter between tube blocks, limited direct contact of the cooling tubes with the tube blocks.

Soon after startup, aluminum-uranium-235 alloy slugs (later replaced by enriched fuel) were introduced in tubes near the edge of the fueled regions of H and C Reactors to flatten the side-to-side power generation rates. This flattening helped to minimize the amount of distortion in the fringe graphite; however, distortion remained a problem, and it later became necessary to replace the control rods with rods that were designed to operate in distorted channels.

The Third Generation of Hanford Reactors

The precursors for the graphite to be used in KW and KE Reactors were selected in 1951; the primary criterion was that the resultant graphite should have a low rate of expansion when irradiated at low temperatures. The only high-temperature data available was for a very low-fluence irradiation at 400°C,

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i) Giberson, R. C., and W. C. Morgan. 1962. Contraction of Graphite: A Comparison of Laboratory and Production Reactor Data - Part I - B, D, F, H, DR, and C Reactors, HW-74155; and: Giberson, R. C., and W. C. Morgan. 1963. Contraction of Graphite: A Comparison of Laboratory and Production Reactor Data - Part II - KW and KE Reactors, HW-76060.

and the measurements indicated that a small amount of expansion had occurred.[j] As shown in Figure 1, TSGBF fulfills the above criterion; however, it contracts much more rapidly does CSF graphite when irradiated at temperatures above about 300°C. It was later shown (Helm 1966) that, following the period of rapid contraction, TSGBF begins to expand in the transverse direction and loses strength as large cracks develop between the particles.

By 1964, distortion of the graphite core was beginning to interfere with reliable operation of the vertical safety rods (VSRs), and boron-steel balls used in the backup emergency-shutdown system were sometimes lost in the stack after a ball drop (Alexander and Russell 1964). By enlarging holes in the top shield, and boring the VSR channel liners, the VSR channels were lined with high-strength graphite sleeves; a similar renovation program of VSR channels was later instituted at C Reactor.

Control-rod channels were renovated, and tube blocks were overbored, to alleviate problems due to distortion caused by the rapid shrinkage of TSGBF graphite. Shortly before the K Reactors were deactivated, graphite in the central region of both reactors reached maximum density and began to expand in the transverse direction; however, no operational problems occurred as a result of this change in behavior.

The Fourth Generation of Hanford Reactors

N Reactor, the only fourth-generation reactor built at Hanford, was designed to generate electricity, as well as producing special materials. It is a graphite-moderated pressure-tube reactor, of quite different design than the previous three generations of production reactors. The results of tests (Love et al. 1961), to determine the effects of a tube-rupture under full power conditions, convinced the designers to add keys to the moderator blocks. The resultant structure is shown in Figure 6.

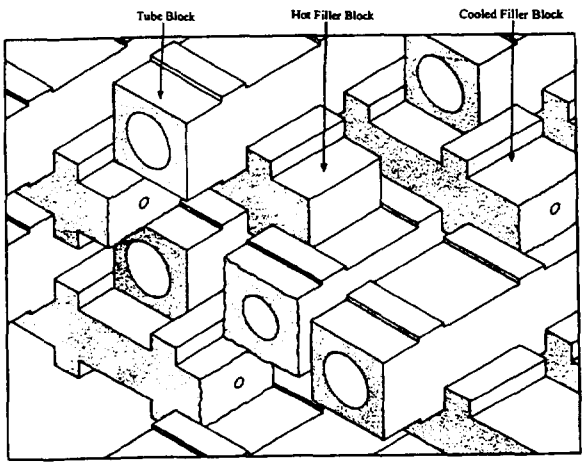

Figure 6. Isometric View of the N Reactor Graphite Blocks.

N Reactor was the first of the Hanford Reactors to benefit from the knowledge that graphite contracts in the transverse direction when irradiated at temperatures above about 300°C. The petroleum coke (Texas-Lockport) selected for grade TSX (N Reactor moderator) graphite, and the increase in graphitization temperature (to 3000°C), were chosen to minimize this shrinkage;[k] however, the chosen coke yielded a very anisotropic graphite, which contracts much more rapidly than grade CSF in the parallel direction under irradiation. This shrinkage in the parallel direction caused fracturing of the keys after they came in contact with the adjacent moderator bars; the tall keys tended to break out of the filler bars, while the short keys (on the tube blocks) tended to shear off at a 45-degree angle.[l] After the short keys had sheared off, the resultant surface acted as a ramp to lift and separate adjacent moderator bars; thus, the "ramp effect" partially compensated for the transverse shrinkage of the moderator bars.

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j) Riley, W. C. 1952. Evaluation of Texas Coke Graphite as a Pile Material, HW-26541.

k) Davidson, J. M., E. M. Woodruff, and H. H. Yoshikawa. 1960. Technical Basis for NPR Graphite Recommendation, HW-64287.

l) Morgan, W. C. 1966. N-Reactor Stack Integrity, BNWL-CC-455.

Before N Reactor was deactivated, however, the graphite in the central region had ceased to contract in the transverse direction, and had begun to expand at an accelerating rate. This expansion, combined with the continued parallel contraction and increased rate of ramping would soon have closed the gap between the stack and the top shield. Studies had been undertaken, and several promising approaches had been devised, to prevent contact of the stack with the shield; however, before any of these measure could be implemented, the decision was made to deactivate N Reactor.

A decision has not yet been reached on the ultimate disposal of N Reactor; however, the moderators of the other eight production reactors will each be transported (as a unit, still encased in the reactor shielding) from their existing sites, near the Columbia River, to higher ground. They will then be buried in a massive pit, prepared especially for their disposal near the center of the Hanford site.

Table 3. Summary of Important Events in the History of the Hanford Reactors

Date	Event
Sept 1944	B Reactor started up; xenon-135 problem discovered.
Feb. 1945	B Reactor attained design power; F Reactor started up.
Late 1945	Discovery that center of top shields at B and D Reactors were bulging.
Mar. 1946	B Reactor placed on standby status (until July 1948).
Oct. 1947	Began increasing power levels at D and F Reactors.
Jan. 1948	Carbon dioxide added to the (He) blanket gas at D Reactor.
Fall 1949	Central zone expansion halted or reversed at B, D, and F Reactors; H Reactor started up at 100% CO_2 and graphite temperatures above 300°C in the central zone.
June 1952	First measurements of transverse contraction in graphite; this provided confirmation of the reason for the subsidence of the central moderator at H Reactor to below lay up level.
Nov. 1952	Startup of C Reactor; bore diameter of tube blocks varied to attain more uniform temperature distribution, and trunion blocks used to support tubes.
Jan. 1955	Startup of KW Reactor; graphite was selected (in 1951) for minimum low-temperature expansion rate.
Oct. 1958	Suggested design changes for N Reactor to reduce maximum graphite temperatures.
Jun. 1964	Startup of N Reactor; graphite was selected (in 1960) for minimum high-temperature contraction rate.
Feb. 1970	KW Reactor shutdown following measurements confirming reversal of contraction for the graphite in the central zone.
Mid-1970s	Reversal of contraction in central zone of N Reactor; exact date is obscured by effect of filler-block keys overriding tube blocks.
Jan. 1987	N Reactor placed on standby status.

Summary

The nuclear grade graphites used in the four generations of Hanford Production Reactors were all selected with only little information regarding the changes in dimensions and physical properties that would be produced by neutron radiation under reactor operating conditions. Each generation of production reactors was constructed (primarily) from a new grade of graphite, and was designed to operate under different conditions than was the previous generation. The lack of data regarding the effects of neutron radiation under the, ever changing, operating conditions presented challenges to reactor operations staff and the scientific-support staff, as the effects of neutron radiation manifested itself through the changes in dimensions and physical properties of the graphite moderator bars. Table 3 contains a brief summary of important events in the history of the Hanford Production Reactors.

Later, in-depth studies by scientists at Hanford and at other research establishments greatly expanded our knowledge regarding radiation-induced changes in the chemical and physical properties of graphites, the effects of manufacturing variables (especially precursor materials) on the magnitude of the changes, and the importance of reactor design and operational variables that can minimize the impact of these changes on reactor operations. However, at the Hanford Production Reactors, knowledge regarding the effects of neutron radiation on graphite was gained by observation of the effects of those changes on the operational characteristics of the reactors, the physical distortions of the graphite moderator blocks, and the physical properties and chemical reactivities of small samples of graphite that had been exposed to neutron radiation within the reactors themselves.

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