

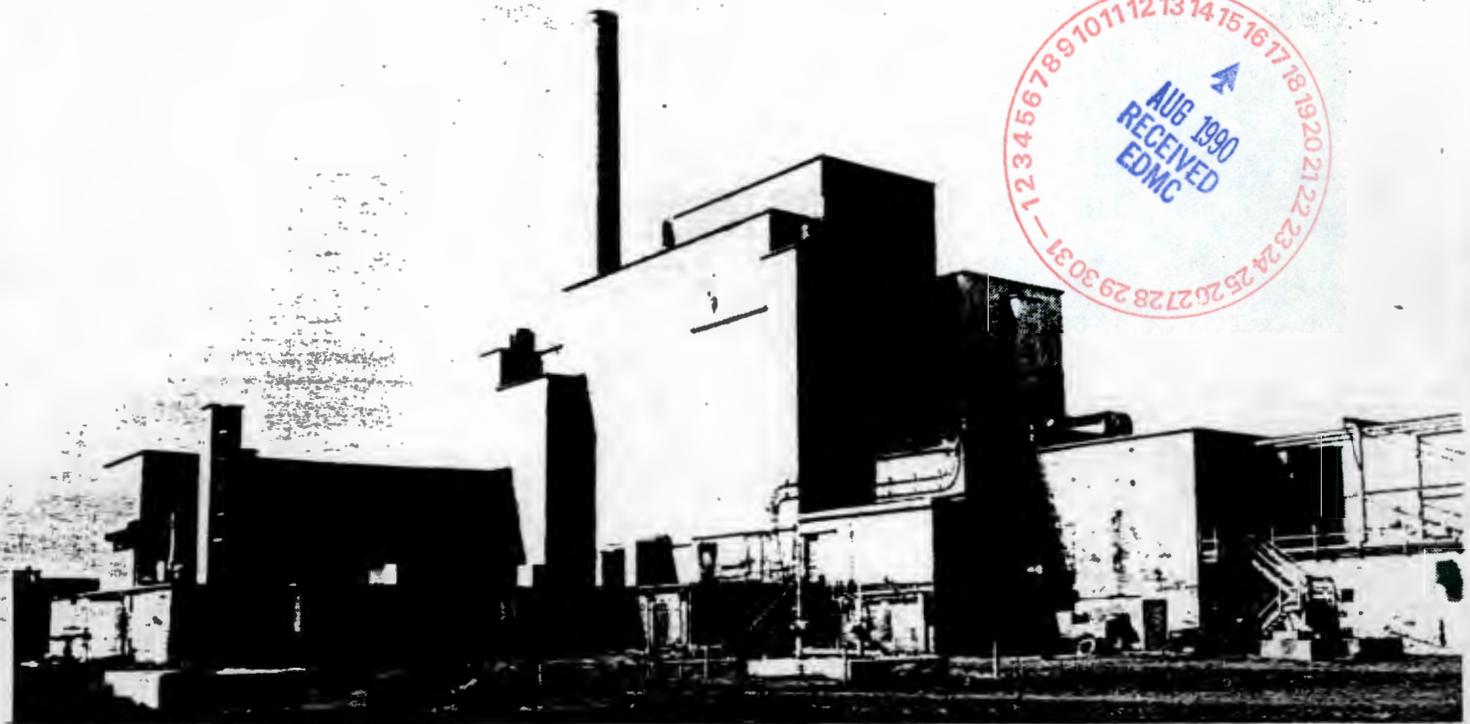
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## RADIONUCLIDE INVENTORY AND SOURCE TERMS FOR THE SURPLUS PRODUCTION REACTORS AT HANFORD



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EDITED BY:

R.L. MILLER AND J.M. STEFFES

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SUMMARY

Radionuclide inventories have been estimated for the eight surplus production reactors at Hanford. The inventories listed represent more than 95% of the total curie burden; the remaining 5% is distributed in piping, tunnels, and various other locations within the reactor building and unaccounted for inventories within the reactors or fuel storage basins. Estimates are conservative as the methodology was designed to overestimate the radionuclide inventories in the facilities.

The estimated inventory per reactor facility ranges from 13,000 curies to 58,000 curies. The majority of the present inventory consists of tritium, carbon-14, cobalt-60, and nickel-63.

The information in this document combines data from past characterization efforts and introduces adjustments for added information and refinement.

Since the reactors have been shut down from 15 to 20 years, many of the shorter half-life radionuclides have decayed to insignificant levels and are therefore not addressed. Trace amounts of some longer half-life radionuclides are listed only to show that they were evaluated.

The inventory of hazardous materials in the reactor facilities is also addressed.

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ACKNOWLEDGEMENTS

The editors wish to acknowledge the contributions of J. A. Adams, J. A. Hall, and M. L. Smith, who prepared the first draft and provided additional data and calculations for this final document.

Valuable comments were provided by Hanford Engineering Development Laboratory personnel as part of their independent technical review of the final draft of this document.

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## 1.0 INTRODUCTION

### 1.1 PURPOSE

The purpose of this document is to provide estimated inventories of radionuclides and other hazardous materials in the eight Hanford 100 Area surplus production reactor buildings. This information is intended to support the preparation of an Environmental Impact Statement (EIS) currently being prepared by Battelle Pacific Northwest Laboratory (PNL) for the final decommissioning of these facilities.

### 1.2 SCOPE

The estimated reactor radionuclide inventories are based on previous analysis and physical measurements. Supporting documentation is included in the appendices and references. Hazardous material inventories are based upon recent walk-through inspections of each facility.

For this document, each reactor facility was grouped into three sections: 1) the reactor block, which includes the graphite moderator stack, biological and thermal shields, process tubes, and the safety and control systems; 2) the irradiated fuel storage basin; and 3) contaminated portions of the facility that are outside the reactor block and fuel storage basin but are within the reactor building decommissioning boundary.

Where numerical data contained in this report were estimated, the estimates are conservative. They were designed to overestimate the radionuclide and hazardous material inventories present. Impacts of special tests, special fuel loadings, special control rods, etc., were not calculated in the inventory estimates since their effect on the overall inventory is not considered significant. For the same reason, no attempt was made to examine the spatial effects that occur due to fuel, control rods, test holes, special loadings, etc. Prior to actual decommissioning a detailed characterization of

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each reactor facility will be performed as necessary to provide definitive estimates of waste volumes, radiation dose rates, and radionuclide inventories.

At the time the reactor decommissioning planning documents were prepared, the date of March 1, 1985, was selected as a target for beginning decommissioning work. The date has been maintained in this document for decay corrections to inventory figures.

All radionuclide inventory values are expressed in curies of the particular isotopes present. Readers are advised that even though values for some radionuclides are in the thousands of curies while others are extremely small, in some instances the small value is more important environmentally than the large value, due to such factors as half-life period and biological effects. This is especially true for transuranic radionuclides, which have very large curie to rem (roentgen-equivalent-man) conversion factors.

### 1.3 HISTORY OF HANFORD 100 AREA REACTORS

The Hanford Site (Figure 1) occupies 540 square miles in southeastern Washington State and was commissioned in 1942 for the production of plutonium by the Manhattan Engineering District of the U.S. Army Corps of Engineers. Eight graphite-moderated reactors and associated support facilities were constructed in the Hanford 100 Area between 1942 and 1954 to support the national defense plutonium production effort. The reactors were designated B, C, D, DR, F, H, KE, and KW. The reactors were shut down between 1964 and 1971. Table 1 summarizes the reactors' operating histories.

A ninth production reactor, N Reactor, was started up in 1963 and is still in operation. Not considered for decommissioning at this time, N Reactor is not included in this report.

During the operating life of the reactors, several changes were made in the hardware and fuel loadings to support operation at higher power levels and greater production efficiency. These changes have significantly influenced

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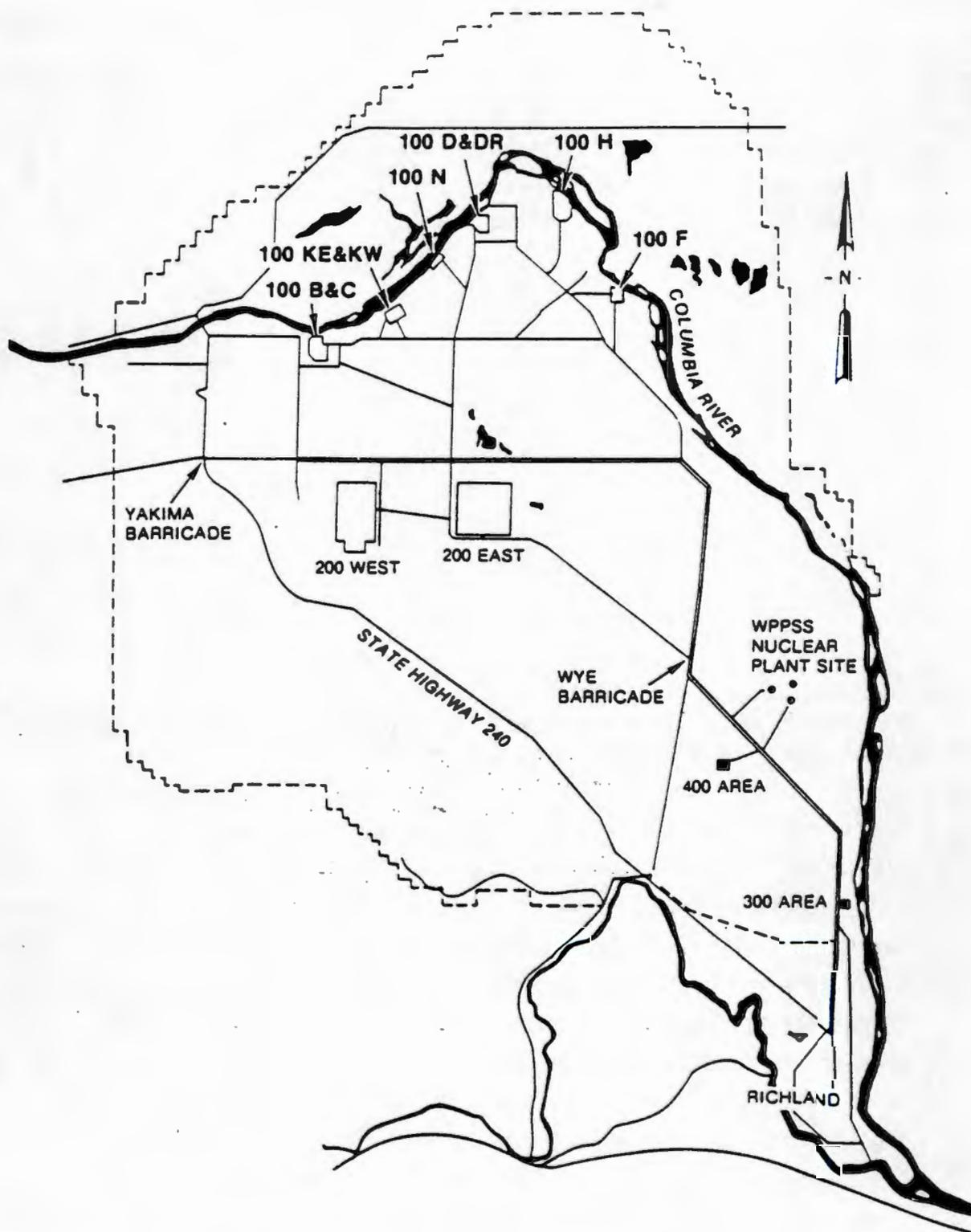


Figure 1. Hanford Site. The eight production reactors are situated along the Columbia River.

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TABLE 1

REACTOR OPERATING HISTORIES

<u>Reactor Facility</u>	<u>Initial Startup Date</u>	<u>Final Shutdown Date</u>	<u>Years Operated</u>
B	09/26/44	02/13/68	22*
C	11/18/52	04/25/69	17
D	12/17/44	06/26/67	23
DR	10/03/50	12/30/64	14
F	02/25/45	06/25/65	20
H	10/29/49	04/21/65	16
KE	04/17/55	01/28/71	16
KW	01/04/55	02/01/70	15

---

\*B Reactor was shut down and held in standby status from 03/19/46 to 06/02/48, then restarted and operated until February 1968.

the quantities and distribution of radionuclides remaining in the reactors. Higher equilibrium power levels in the older reactors were achieved during the early 1950's by using various control rod patterns and installing neutron-absorbing material in some of the process tubes in place of fuel. At the same time, some enriched fuel loadings (higher uranium-235 concentration) were introduced to increase the flattening or evening-out of the power levels. Then in the late 1950's even more dramatic power level increases were achieved in the six older reactors by introducing an "enrichment ring" and significantly increasing the cooling water flow rate through the reactors. The enrichment ring consisted of several hundred enriched fuel columns installed in a ring pattern near the outside of the process tube pattern. The effect was to create a central zone of the reactor that had nearly equal individual tube powers. Prior to loading the enrichment rings, the power

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level of the reactor was determined by the "hottest" tubes, which were usually located near the center of the reactor. With the flattened power distribution, the limiting condition for the older reactors became the bulk outlet coolant temperature for the entire reactor.

The overall effect of flattening the power in the central zone of the reactor and increasing the cooling water flow resulted in final equilibrium power levels in the 2000 MW range for the older reactors, which had design power levels of 250-350 MW, and a final level of 4400 MW for the K reactors, which had design power levels of 2500 MW.

#### 1.4 DOCUMENT ORGANIZATION

The major portion of the document is divided into three sections corresponding to the groupings of a reactor facility: the reactor block, the irradiated fuel storage basin, and the other areas of the building that might contain some radionuclides. Within each of the three sections, the methods and sources of characterization data, the calculation adjustments and assumptions, and the inventory estimates are presented.

The information in this document combines data from past characterization efforts and introduces adjustments for added information and refinement.

In order to determine a conservative and realistic radionuclide inventory estimate, several general assumptions were made to simplify the complexity of the materials and conditions in the reactors. These simplifying assumptions and the adjustments, along with a rationale, are provided in the section under the reactor divisions. In all cases, conservative assumptions and adjustments were applied.

The radionuclide inventories were estimated for the major components of the surplus reactors. An attempt was made to provide two significant figures for each estimate. In some cases the uncertainty involved in making the estimate

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precluded the use of the two significant figures. An example of the uncertainty is the calculation of trace nuclides in the shields where the parent nuclide concentration was estimated from handbook values for "typical" materials.

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## 2.0 REACTOR BLOCK

The reactor block (Figure 2) is located near the center of the reactor building. Horizontal control rod penetrations are on the left side of the block (when facing the reactor front face), experimental test penetrations are on the right side. Fuel discharge and storage areas are located directly behind the rear of the reactor. The vertical safety rod and ball 3X system penetrations are on the top of the reactor. The ball 3X system is an emergency shutdown system consisting of neutron-absorbing balls that could be released into the reactor core from hoppers above the reactor.

A typical older reactor block consists of a graphite moderator stack encased in a cast iron thermal shield and a biological shield, consisting of alternating layers of Masonite and steel (Figures 3 and 4). In both of the K reactors, heavy-aggregate concrete was used as a biological shield. The reactor block rests on a massive concrete foundation.

The principal components of a production reactor block are:

- o The reactor moderator stack, which is an assembly of graphite blocks cored to provide channels for process tubes, control rods, and other equipment.
- o Process tubes, which contained the uranium fuel elements and provided channels for cooling water flow.
- o Horizontal control rods.
- o Vertical safety rods.
- o Ball 3X system, for dropping neutron-absorbing balls into vertical safety rod channels for emergency reactor shutdown.
- o Monitoring and experimental test equipment.
- o Thermal and biological shielding, surrounded by a heavy, vault-like steel outer shell equipped with gas-tight seals for the reactor block penetrations.

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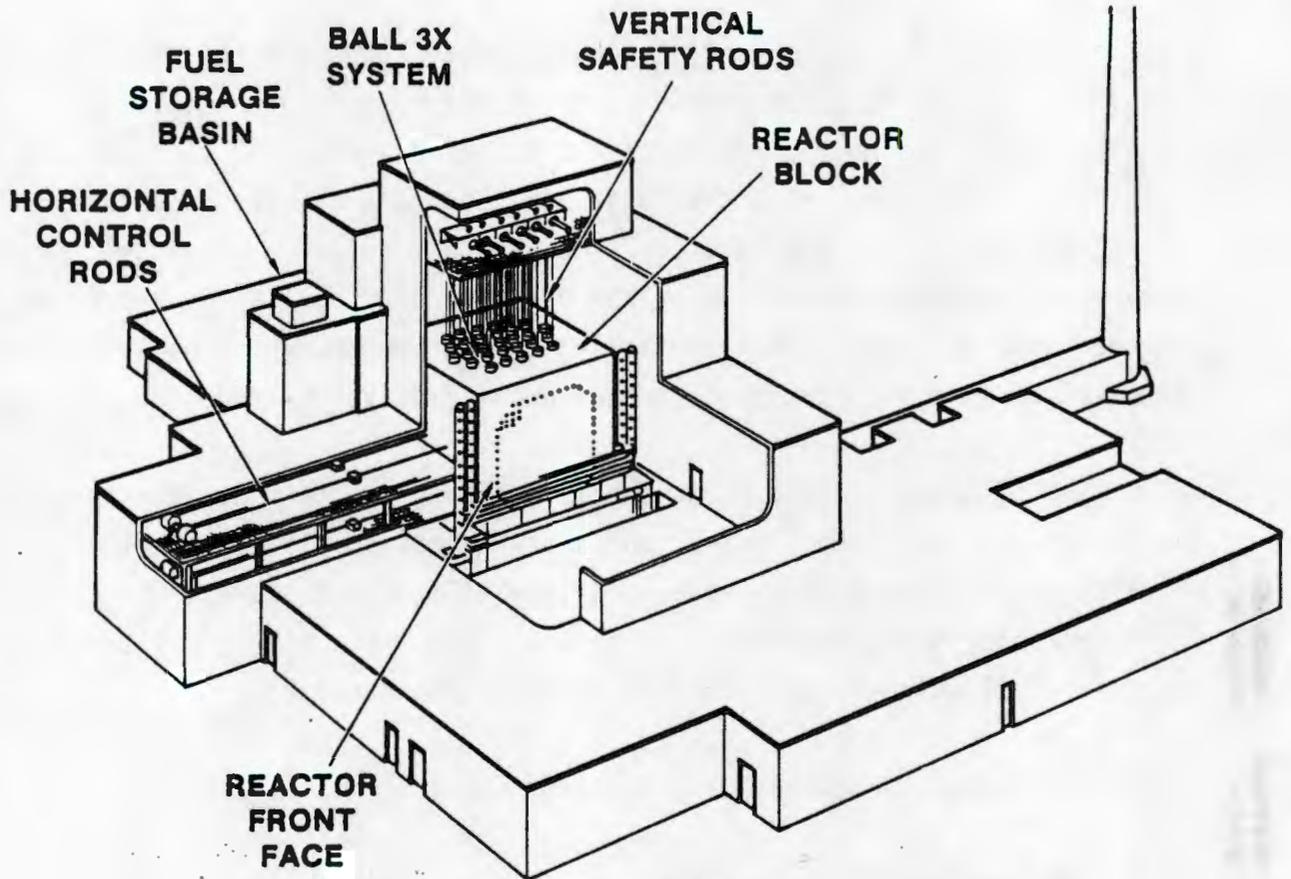


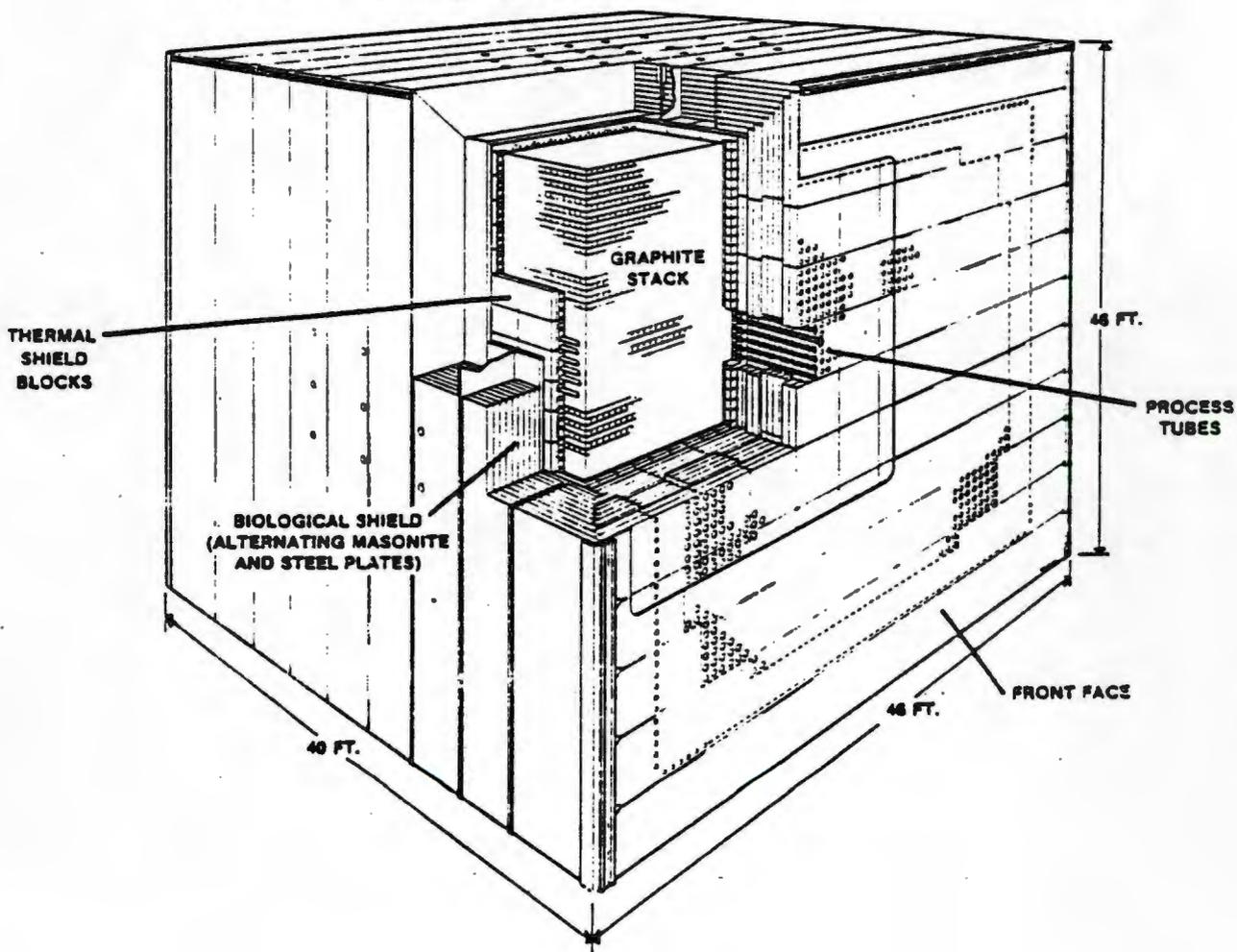
Figure 2. Typical Reactor Facility.

For inventory purposes, the reactors are assigned to two groups: 1) the older reactors B, C, D, DR, F, and H; and 2) the newer KE and KW reactors. The older reactors' thermal shields and graphite stacks are of similar size and were constructed of similar materials. The older reactors also had similar neutron flux distributions.

The two K reactors are comparable to the older reactors, having been constructed from similar drawings and specifications. But they had graphite stacks 1-1/2 times larger than the older reactors, larger fuel elements, more

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# REACTOR BLOCK



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Figure 3. Reactor Block Construction.

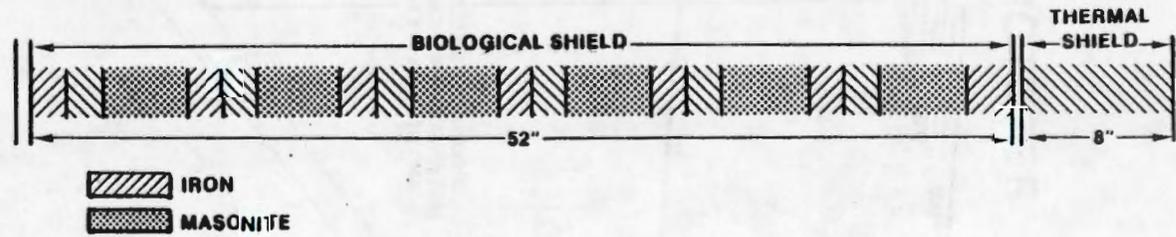


Figure 4. Cross Section of Biological and Thermal Shield.

and different-material process tubes, and heavy-aggregate concrete biological shields. Additionally, the two K reactors were operated at a higher power level. Table 2 shows the comparison of the reactor groups.

TABLE 2  
HANFORD PRODUCTION REACTOR DESIGN DATA

Reactors	Graphite Stack Dimensions (ft)			Process Tubes			Thermal Shield	Biological Shield	Shield	
	Front to Rear	Top to Bottom	Side to Side	Number	Type	ID (in.)	Type	Thick-ness (in.)	Type	Thick-ness (in.)
B, C,* D, DR, F, H	28	36	36	2004	Aluminum	1.75	Cast Iron	8-10	Steel and Masonite	52
KE, KW	33.5	41	41	3220	Zircaloy and Aluminum	1.8	Cast Iron	10	Heavy-Aggregate Concrete	45-83

\*C Reactor has slightly larger diameter process tubes than the other reactors in this group. It has a heavy-aggregate concrete top biological shield (84 in. thick) in place of steel and Masonite.

## 2.1 METHODS AND SOURCES FOR REACTOR BLOCKS

The main source of characterization data for the reactors is recorded in the DR Reactor Characterization (Appendix A). The radionuclide concentration data in Appendix A are used in this document to derive estimated inventories of radionuclides for the DR Reactor. Inventory numbers for the other older reactors (B, C, D, F, and H) are based on the characterization results from the core sampling of the DR Reactor block, then adjusted for operating history and fluence differences. Inventory numbers for the K Reactors were based both on some sample analyses and by adjusting the DR Reactor data.

9 3 1 2 8 7 2 0 4 2 2

Where sample analysis data were not available or did not appear adequate for the specific component, a calculation was made using basic activation computational techniques or using the data set presented in Reference 1.

## 2.2 CALCULATION ADJUSTMENTS AND ASSUMPTIONS FOR REACTOR BLOCK

The principal components of each reactor block are the graphite moderator stack, the cast iron thermal shields, the process tubes (which contained the fuel charges); the control rods, safety rods, emergency shutdown ball 3X system, and the biological shields. Each of these components is addressed separately in paragraphs 2.2.1 through 2.2.5.

### 2.2.1 Graphite Moderator Stack

The graphite moderator stack consists of 4-foot-long graphite blocks stacked to provide a central region for fuel loading and an outer region for a neutron reflector. Each of the older reactors (B, C, D, DR, F, and H) contains 2004 process tube openings; the two K Reactors each have 3220 process tube openings. In addition to these openings, the graphite block has openings for control rods, safety rods, test facilities, and instrumentation. The original four Hanford reactors (B, D, DR, and F) are very nearly identical; the DR Reactor has an additional test facility. The C and H Reactors have more control rods (15 versus 9), more safety rods (45 versus 29) and more test facilities (13 at C Reactor, 10 at H Reactor, 3 each at B, D, F, and 4 at DR). Each K Reactor has 20 control rods, 45 safety rods, and 16 test facility openings.

The openings provided for the control rods, safety rods, and test facilities are a small fraction of the total stack volume. The radionuclide inventory of the graphite is based on the mass of the graphite. By using the graphite volume of a B, D, or F Reactor, a small conservatism is provided for the other older reactors.

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As discussed in Section 1.3, at the end of their operating life, each reactor had an enrichment ring installed to provide power flattening in the central zone of the reactor. It should be noted that the flattening of power in the central zone affected the neutron flux levels only in the radial direction from an axis in the front-to-rear direction. The axial flux distribution was not flattened except to a very limited extent achieved by judicious placement of control rods. The axial flux distribution was closely approximated by a cosine distribution in the front-to-rear direction. This cosine shape of the flux allowed another convenient means of applying a conservative factor, i.e., assume that the flux level is constant at the peak level over the entire fuel charge length. This factor of conservatism was used whenever a calculation was used in place of sample data.

An additional conservative adjustment was made in the radial direction. Although the enrichment ring was several lattice units (process tubes) in from the outside of the charge pattern, for calculation purposes the flattened zone was assumed to extend to the entire active region of the reactor.

Table 3 summarizes the characteristics used for the reactor graphite stacks.

Radionuclide inventories in the graphite stack were determined for the DR Reactor by using the average concentration values presented in Appendix A, decay-correcting to March 1, 1985, and multiplying by the volume-adjusted stack mass. A similar procedure was used for the K reactors' inventories using KW sample analysis data (Appendix B).

The estimated radionuclide inventories for the old reactors, other than DR, were calculated by taking the ratio of the neutron fluences or the total effective full-power operating days as a multiplier for the DR Reactor data. The values obtained were decay-corrected to March 1, 1985. Table 4 shows the equivalent full-power days operated, the fluence, and the decay times for each reactor.

TABLE 3

REACTOR STACK PHYSICAL CHARACTERISTICS

<u>Parameter</u>	<u>Old Reactors</u>	<u>K Reactors</u>
Dimensions of Stack:		
Side to Side	36 ft (11 m)	41 ft (12.5 m)
Top to Bottom	36 ft (11 m)	41 ft (12.5 m)
Front to Rear	28 ft (8.5 m)	33.5 ft (10.2 m)
Fuel charge length	23.8 ft (7.2 m)	28.1 ft (8.6 m)
Number of process tubes (fuel charges)	2004	3220
Lattice spacing for process tubes (fuel charges)	8-3/8 x 8-3/8 in.	7-1/2 x 7-1/2 in.
Volumes:		
Entire stack	1028 m <sup>3</sup>	1595 m <sup>3</sup>
Active zone	658 m <sup>3</sup>	1001 m <sup>3</sup>
Reflector	370 m <sup>3</sup>	594 m <sup>3</sup>
Process tubes	(24 m <sup>3</sup> )*	(45.3 m <sup>3</sup> )*
Control and Safety rods	(4.1 m <sup>3</sup> )*	(4.8 m <sup>3</sup> )*
Test facilities	(0.3 m <sup>3</sup> )*	(1.4 m <sup>3</sup> )*
Density of graphite	1.7 g/cm <sup>3</sup>	1.7 g/cm <sup>3</sup>
Mass of graphite:		
Active zone	1.07 x 10 <sup>9</sup> g	1.61 x 10 <sup>9</sup> g
Reflector	6.3 x 10 <sup>8</sup> g	1 x 10 <sup>9</sup> g
Flux levels (neutrons/cm <sup>2</sup> -s)		
Active zone	5 x 10 <sup>13</sup> /cm <sup>2</sup> -s	1 x 10 <sup>14</sup> /cm <sup>2</sup> -s
Reflector (center)	5 x 10 <sup>12</sup> /cm <sup>2</sup> -s	5 x 10 <sup>12</sup> /cm <sup>2</sup> -s
(outer edge)	1 x 10 <sup>12</sup> /cm <sup>2</sup> -s	1 x 10 <sup>12</sup> /cm <sup>2</sup> -s

\*Volume of openings not included in entire stack volume calculation.

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TABLE 4

OPERATING DATA AND DECAY TIMES FOR RADIONUCLIDE CALCULATIONS

<u>Reactor</u>	<u>Final Operating Power Level (MW)</u>	<u>Full-Power Days Operated</u>	<u>Fluence,* 10<sup>22</sup> per cm<sup>2</sup></u>	<u>Decay time to 3/1/85 (yrs)</u>
B	1960	5092	2.20	17
C	2310	5108	2.21	15.8
D	2005	4933	2.13	17.7
DR	1925	3610	1.56	20.2
F	1935	4181	1.81	19.7
H	1955	3990	1.72	19.9
KE	4400	5311	5.51	14.1
KW	4400	5043	5.23	15.1

\*The peak neutron flux assumed was  $1 \times 10^{14}/\text{cm}^2\text{-s}$  for the K Reactors and  $5 \times 10^{13}/\text{cm}^2\text{-s}$  for all other reactors.

### 2.2.2 Thermal Shield

A cast iron thermal shield surrounds the graphite stack. Constructed of overlapping blocks, an older reactor's shield varies in thickness (8 to 10 inches) on the sides, top, bottom, front and rear. The K Reactor thermal shields are all 10 in. thick. The inventory for the thermal shield also includes the contribution from the gunbarrels, which are steel tube connections for cooling water and instrumentation. Figure 5 is a simplified cross-section of a process tube penetration through the reactor, showing the gunbarrel, process tube, thermal and biological shields, and graphite relationships.

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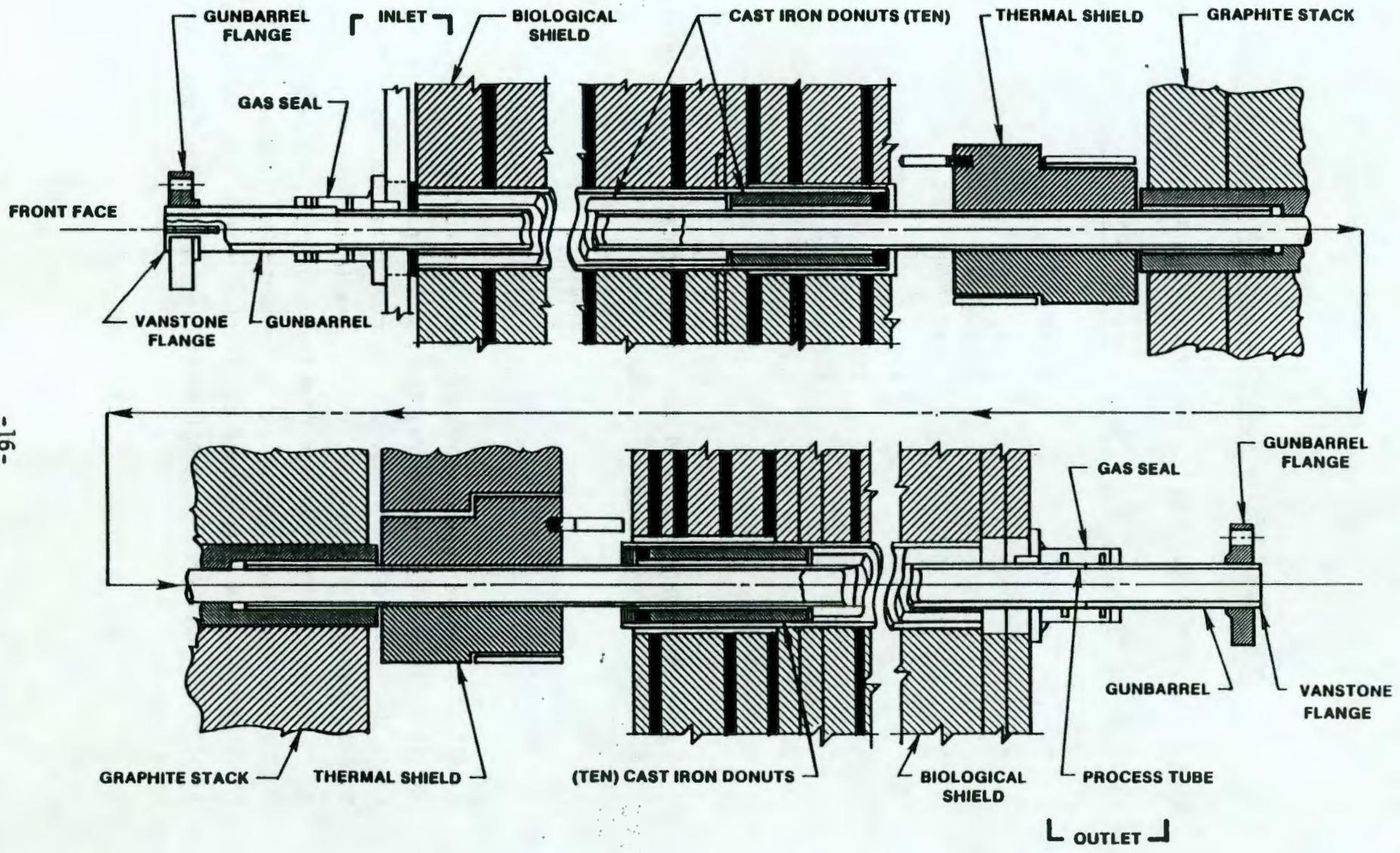


Figure 5. Process Tube Channel Cross Section.

In addition to the radionuclide analysis of the DR Reactor thermal shield samples provided in Appendix A, a chemical analysis was performed on samples obtained from a mock-up which had been assembled using original reactor components serial-numbered for traceability. A thermal shield cast iron block and a gunbarrel were analyzed. Their chemical compositions are given in Table 5. The chemical analysis focused on determining parent or target nuclei of long-lived radionuclides; therefore, the cobalt with its short (5-year) half-life was not included in the analysis.

TABLE 5

CHEMICAL COMPOSITION OF REACTOR PARTS

<u>Chemical Element</u>	<u>Cast Iron from the Thermal Shield Block</u>	<u>Sample of Steel from the Gunbarrel Tip</u>
Ti	0.012%	0.012%
V	0.004%	0.004%
Cr	0.156%	0.042%
Mn	0.248%	0.113%
Fe	98.8%	99.4%
Ni	140 ppm	130 ppm
Cu	0.288%	0.071%
Zn	0.009%	0.006%
Ga	0.002%	0.002%
Ge	0.004%	0.002%
Se	0.001%*	0.001%*
Sr	0.001%*	0.001%*
Y	0.001%*	0.001%*
Zr	0.004%	0.0
Nb	4.9 ppm	2.6 ppm
Mo	210 ppm	39 ppm

\*Minimum detection limit.

Stainless steel (18-8), 3/4-in., Schedule 40 tubes were embedded front-to-rear in the side, top, and bottom thermal shields to provide cooling water for the shields. The front and rear shields were cooled by the process tubes. The induced activity of the thermal shield cooling tubes was calculated based on the fluence level and standard material specifications for 18-8 stainless steel. The calculated inventories for nickel-63, nickel-59, cobalt-60, molybdenum-93, and niobium-94 in the cooling tubes are included in the thermal shield inventories.

To account for the activity of the gunbarrels, the assumption was made that the inventory in the gunbarrels would be less than the thermal shield portion that they replace. This assumption is conservative for the radionuclides of interest as can be seen by inspection of the nickel, zirconium, niobium, and molybdenum composition values presented in Table 5.

The active portion of the thermal shields was considered to have the same length and height as the graphite stack. This assumption neglects the "corners" at each edge of the total shield; however, the neutron flux level in these corners is very low compared to the central portion of each shield face.

The inventory estimates for the thermal shields contain a conservatism similar to the graphite stack inventories. The concentrations, whether from measurements or calculated data, were applied to the entire active portion of the shield with no adjustment for lower neutron flux levels near the edges. The general method of determining the cobalt-60, nickel-63, and nickel-59 inventories was to consider the shields as eighteen slabs, each 1/4 in. thick. The measured values, as a function of distance into the shield from the DR Reactor data in Appendix A, were plotted on graph paper. From the graph a concentration for the desired radionuclide was determined for any specific 1/4-in. slab. By multiplying the concentration by the mass of the slab, the curies of a nuclide in each slab were determined. The sum of the curies over all the slabs gives the total shield inventory.

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The estimated trace radionuclide inventories were calculated using a flux level of  $10^{11}/\text{cm}^2\text{-s}$  for the effective full-power days of operation and the impurity levels from the chemical analysis.

### 2.2.3 Process Tubes

The six older reactors used process tubes made of 1100 aluminum alloy. The chemical composition specification required a minimum of 99% aluminum. Other elements were allowed only to the maximum amounts listed in Table 6. These impurities do not generate significant quantities of radionuclides with long half-lives that are of concern. Gamma radiation spectrum measurement of aluminum samples that were present in the K Reactors for the entire operating life of the plant did not indicate measurable concentrations of aluminum-26.

TABLE 6

#### ALUMINUM ALLOY PROCESS TUBE IMPURITIES

<u>Element</u>	<u>Maximum Amount</u>
Cu	0.20%
Si + Fe	1.00%
Zn	0.10%
Mn	0.05%
Li	0.008%
Cd	0.003%
B + Co	0.001%
Others, each	0.05%

About 73% of the process tubes in the K Reactors are made of Zircaloy-2 and had been in use for 7 years prior to final shutdown. A chemical analysis of a Zircaloy-2 process tube was made to determine the possible elements that would produce long half-life nuclides. The analysis indicated 416 ppm nickel, 99.5% zirconium, and 14 ppm niobium. The only other element of concern is

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molybdenum, which is controlled by the purchase specification to be less than 50 ppm. Recent vendor reports for Zircaloy-2 purchased at Hanford indicate 25 ppm (minimum detectable) from Western Zirconium and 10 ppm (minimum detectable) from Wah Chang. To be conservative a value of 50 ppm was used for molybdenum.

The cladding used for the fuel elements contained 1% nickel, which combined with impurities in the cooling water to become part of the corrosion film inside the tube. A sample analysis was performed for both the film and the aluminum process tube material for several tubes at the DR Reactor (Appendix A). A comparable corrosion film analysis was not performed for the K Reactors because the activity of the K Reactor film is not expected to be significantly different since the same process water chemistry was used at all the reactors.

#### 2.2.4 Reactor Control System

The reactor control system included the control rods, safety rods, and ball 3X system. The control rods, moved into and out of passages in the graphite core, controlled startup transients and power level during equilibrium operation. During later years, as the graphite stacks became distorted by growth and shrinkage, the control rod channels became distorted. Several "jointed" type rods were tested to allow the rod to follow the curved path of the channel. Also some testing was made of uncooled rods. All the original control rods were water cooled. No attempt was made to track the history of control rod replacement or short-time use of exotic designs.

The safety rods were located on top of the reactor. Electromagnets held the rods with just the rod tips in the top thermal shield. Only on reactor shutdown, automatic or intentional, were the safety rods dropped into the reactor proper. As the name implies, the safety rods were only a backup to the control rods. The tips of the safety rods became activated due to neutron streaming through the safety rod channels in the graphite stacks.

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To back up the safety rods, a separate emergency shut-down system was provided. This was the ball 3X system, comprised of hoppers full of small boron-steel balls that would automatically drain into the safety rod channels if the safety rods did not terminate the chain reaction after receiving a shut-down signal. Throughout the operating history of the Hanford reactors, the ball 3X systems were never required as the primary shut-down mechanism. However, the ball 3X systems were routinely tested manually to assure that all the balls in the hoppers would drain into the channels in the graphite stack. During the later years of operation as cracks and general shifting of the graphite stacks formed holes and ledges along the channels, balls would become trapped in the stack. These trapped balls caused a loss of reactivity which was overcome by adding more enriched fuel. The amount of enrichment needed to overcome the trapped balls was compared to the mass of balls in the reactor. For the DR Reactor, this equated to 72 pounds of balls. Measured data on the gamma radiation of a small number of irradiated balls were used to compute the amount of cobalt-60 that would be present in the trapped balls. Using this method, a cobalt-60 inventory of 110 curies was computed for the balls remaining in the graphite stack at the DR Reactor.

The inventory of the control rods and safety rod tips is assumed to be the same as the inventory of the thermal shield section (opening) that they replaced. Hence, the inventory of the rods are "included" in the thermal shield estimates.

#### 2.2.5 Biological Shield

The biological shield surrounds the thermal shield. The biological shields for the six older reactors were made of alternating laminated layers of steel and Masonite. As the single exception of the older reactors, the C Reactor has a 7-foot-thick, heavy-aggregate concrete top shield. Both K Reactors have heavy-aggregate concrete biological shields. While not especially intended as shields, all the reactors have massive concrete foundations.

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Even though the biological shields are quite large in size and mass, they contain very little of the radionuclide inventory. The data presented in Appendix A indicate that less than 1% of total inventory of the thermal and biological shields combined is in the biological shield. Furthermore, of the inventory in the biological shields nearly all is in the first layer of steel. Considering the accuracy of the data in this document, the contribution of the biological shield cobalt-60, nickel-63, and nickel-59 may be treated as zero.

The amount of calcium-41 was calculated for the foundation and for the K Reactor shields for completeness. The calcium-41 was based on a calcium-40 level of 72,000 ppm for concrete and a flux level of  $10^{10}/\text{cm}^2\text{-s}$ .

### 2.3 ESTIMATES FOR REACTOR BLOCKS

The inventory for the reactor blocks is composed of estimates from the major components of the block: the graphite moderator stack, the thermal shield, the process tubes, and the control system, which includes the control rods, safety rods, and ball 3X systems. The inventory estimates will be tabulated at the end of the sections discussing the individual radionuclides.

#### 2.3.1 Graphite Moderator Stack

Radionuclides in the graphite originate from the carbon, the cover gas, the cooling water (due to occasional tube leaks), and from the impurities present in the coke when it was processed into graphite blocks. The percentages of impurities present in the petroleum when it was baked into coke remained as the coke was further processed into graphite. The amount and configuration of impurities dictated the grade of graphite produced and its eventual suitability for reactor use. Initially the graphite was purified by heat only. Later, a chemical purification process was used to further reduce unwanted elements. The concentrations of radionuclides remaining in the graphite at each reactor reflect the source of the graphite, the grade of

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graphite, and the history of operation. Tritium constitutes about half of the radioactivity present in the graphite. The other half is mainly carbon-14, nickel-63, cobalt-60, and chlorine-36.

#### 2.3.1.1 Tritium

The amount of nitrogen-14 available as a target material in the graphite blocks was dictated by the concentrations in the original coke. The nitrogen-14 captures neutrons and becomes carbon-12 plus tritium. This is the dominant production reaction for tritium in the graphite stack (Appendix D). Tritium has a half-life of 12.3 years and emits low-energy beta particles.

Using the average value for tritium concentrations, shown in Table A-7 of Appendix A, yields a value of  $6.8 \times 10^6$  pCi/g for the DR Reactor graphite for March 1977. Applying the central zone and reflector masses, scaling by the lower flux value assumed for the reflector, and, finally, decay-correcting to March 1985, an estimated inventory of 4900 curies of tritium is found for the DR Reactor stack. To determine the tritium inventories for the other old reactors, the ratio of the fluences is applied to the DR Reactor value of  $6.8 \times 10^6$  pCi/g to obtain the other reactors' concentrations 12.2 years after shutdown. (The 12.2 years is the time between final shutdown of DR and the sample analysis time for data in Appendix A.) The value found for the tritium is then decay-corrected to March 1985 to be consistent with the inventory estimates presented in this document.

For the K Reactors a correction for the higher neutron flux level in the active zone, the greater mass of the active zone, and greater mass of the reflector is made in addition to the adjustments above for the older reactors.

No correction was made for the decay of tritium during the longer operating period of the reactors other than DR. This omission provides a conservatism in the estimates.

Table 7 shows the tritium inventory for the graphite stacks.

9 3 1 2 8 7 2 0 4 3 4

### 2.3.1.2 Carbon-14

Carbon-14 originates from both the nitrogen-14 (n, p) reaction and the (n, gamma) transmutation of carbon-13. The carbon-14 concentration in a graphite stack from the (n,p) reaction is predominantly a function of the available nitrogen in the graphite and cover gas. Samples of graphite from KW Reactor's tube-channel 1880 had a carbon-14 concentration similar to samples from DR Reactor (Appendix B). This similarity is probably due to differences in impurity levels, since a larger fraction of the graphite for K Reactors was chemically purified resulting in a lower impurity level. This lower impurity level could have offset the increased size and higher power levels of the K Reactors in determining carbon-14 inventories.

The average concentration of carbon-14 from the DR Reactor sampling (Appendix A) is  $2.8 \times 10^6$  pCi/g for the active region of the reactor. Applying the adjustments discussed in Section 2.2.1, the inventory of carbon-14 for the DR Reactor is estimated to be 2990 for the active region and 176 for the reflector or 3200 curies total. The carbon-14 inventories for the other older reactors were estimated by using the ratios of the fluences compared to the DR Reactor.

The carbon-14 inventories for the K Reactors were based on the analysis data from the KW Reactor (Appendix B). The KE Reactor inventory was obtained by using the ratio of the fluences for KE and KW Reactors.

The carbon-14 inventories for the reactor graphite stacks are shown in Table 7.

### 2.3.1.3 Nickel

The original elements in the coke used to make the graphite influenced the amount of nickel available as a target material in the graphite. Nickel activation resulted in the generation of nickel-63 and nickel-59. Although a thermal purification process reduced the nickel in the original coke used to make the graphite in the older reactors, the nickel remaining in the graphite

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appears to be proportional to the original concentration of the nickel impurity in the petroleum. For the K Reactor graphites, an additional chemical purification process significantly reduced the amount of nickel impurity present in the graphite. Nickel-62 constituted 3.6% of the total nickel present in the older reactor graphite. Upon capture of a neutron, nickel-62 becomes nickel-63 with a 100-year half-life.

Nickel-58 made up 68.3% of the nickel present in older reactor graphite. Neutron capture converts nickel-58 to nickel-59, with an 80,000-year half-life. It has been estimated that there are 0.008 curies of nickel-59 for each curie of nickel-63 (Appendix E). Other naturally occurring isotopes of nickel are of no interest since they do not produce radionuclides with long half-lives.

The average concentration of nickel-63 reported in Appendix A for the DR Reactor analysis is  $8.4 \times 10^4$  pCi/g. Making the same corrections for active region and reflector as indicated for carbon-14 in Section 2.1.1.2, the total estimated inventory of nickel-63 in the DR Reactor graphite stack is 93 curies. Using the ratio of nickel-59 to nickel-63 derived in Appendix E, the estimated inventory of nickel-59 in the DR Reactor graphite stack is about one curie.

The nickel-63 and nickel-59 estimated inventories for the other reactors were determined by multiplying the ratio of the fluences and the average nickel content compared to the DR Reactor by the DR Reactor values above.

The K Reactors contained chemically purified graphite which resulted in a significantly lower nickel content. The nickel-63 and nickel-59 inventory estimates were made by adjusting the DR Reactor data for fluence and multiplying by the ratio of the average nickel content.

Table 7 shows the nickel inventory estimates for the reactor graphite stacks.

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#### 2.3.1.4 Chlorine-36

The amount of chlorine-35 available for transmutation to chlorine-36 depends upon the residual chlorine in the graphite after manufacture. At the time of construction of the early Hanford reactors (B, D, and F), the stacks were segregated by "purity" with respect to neutron absorbency. The higher grades of graphite (less neutron absorption) were placed in the center of the reactor and lower grades toward the outside. By the time the next generation of reactors (DR, C, and H) were constructed, an added process using carbon tetrachloride and freon was being used to purify the graphite during manufacture.

The amount of chlorine-36 is different for each reactor due to the varying amounts of each grade used during construction of the stack (Appendix C). The inventory for each reactor is shown in Table 7.

#### 2.3.1.5 Cobalt-60

The amount of cobalt-60 present in the reactor graphite stacks is dependent upon the original impurity level of cobalt in the graphite. The purification process discussed earlier in Section 2.3.1 influenced the amount of cobalt-60 presently estimated for the graphite stack inventories.

Cobalt-60 has a half-life of 5.3 years, hence decays by several half-lives during the operation of the reactor. The estimated cobalt-60 inventory for the DR Reactor graphite was found by applying the volume adjustments for the active zone and the reflector and decay-correcting the average of the sample data in Appendix A to March 1985. The cobalt-60 inventories for the C and H Reactors were estimated by correcting the DR Reactor data for the difference in operating days and decay times since final shutdown. For the B, D, and F Reactors the same purification factor that was assumed for the nickel (Section 2.3.1.3) was applied, i.e., the B, D, F operation and decay time corrections and a factor of 2 to account for the higher impurity in the original graphite.

Similar corrections were made for the cobalt-60 in the KE and KW graphite stacks. In the case of the K Reactors the impurity correction factor is 0.04 rather than the 2 used for the B, D, and F graphites.

#### 2.3.1.6 Calcium-41

The grade of graphite used to construct the reactor stacks influences the amount of calcium-41 that is produced during reactor operation. To determine the inventory of calcium-41 in each reactor required the same type of analyses performed for the chlorine-36 (Section 2.3.1.4). The computational method was to find the representative concentration of calcium for each zone of each reactor and then use the data set in Reference 1 to determine the calcium-41 produced.

#### 2.3.1.7 Fission Products and Transuranics

For fission products to enter the graphite stack, both a fuel element failure and a simultaneous failure of the process tube are required, which would allow water and fission products to be carried into the graphite. The amount of fission products remaining in the graphite after nine years of decay would add a few curies to the total inventory of the reactor. As indicated in Table 7, the inventory from mixed fission products remaining in the graphite stacks is made up of plutonium-239, strontium-90, and cesium-137. The estimates were calculated from the DR Reactor data (Appendix A) and applied to the other reactors without correction or adjustment.

Table 7 shows the present estimated radionuclide inventory in the reactor graphite stacks.

TABLE 7  
ESTIMATED RADIONUCLIDE INVENTORY  
IN THE REACTOR GRAPHITE  
AS OF MARCH 1, 1985

Radio-nuclide	Reactor Graphite (curies)							
	B	C	D	DR	F	H	KE	KW
<sup>3</sup> H	8300	8900	7700	4900	5800	5500	30000	27000
<sup>14</sup> C	4500	4500	4300	3200	3700	3500	7000	6700
<sup>60</sup> Co	100	60	90	30	70	40	5	5
<sup>63</sup> Ni	180	28	280	95	190	120	11	15
<sup>59</sup> Ni	1	1	2	1	2	1	-	-
<sup>36</sup> Cl	40	85	35	55	35	70	300	270
<sup>239</sup> Pu	1	1	1	1	1	1	1	1
<sup>241</sup> Am	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
<sup>90</sup> Sr	10	10	10	10	10	10	10	10
<sup>137</sup> Cs	30	30	30	30	30	30	30	30
<sup>133</sup> Ba	32	1	34	10	26	11	1	1
<sup>152</sup> Eu	40	40	40	40	40	40	40	40
<sup>154</sup> Eu	20	20	20	20	20	20	20	20
<sup>41</sup> Ca	190	14	150	90	140	54	1	5

### 2.3.2 Thermal Shield

Table 8 gives the DR Reactor distribution of cobalt-60 and nickel-63 in the right side thermal shield as a function of distance from the inner face of the shield. This distribution is representative of all the thermal shield faces. The curies were obtained by multiplying each activity by the mass of the slab,  $4.46 \times 10^6$  grams.

Total curies of cobalt-60, nickel-63 and nickel-59 for the DR Reactor thermal shield sections are shown in Table 9. The table reflects the different masses of the front and rear shields.

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TABLE 8

DISTRIBUTION OF COBALT-60 AND NICKEL-63 IN THE DR REACTOR  
RIGHT SIDE THERMAL SHIELD  
AS OF MARCH 1, 1985

<u>Distance from Inner Face (in.)</u>	<u>Cobalt-60 (pCi/g)</u>	<u>Nickel-63 (pCi/g)</u>	<u>Cobalt-60 (Ci)</u>	<u>Nickel-63 (Ci/cm)</u>
0.25	$3.4 \times 10^7$	$2.4 \times 10^6$	153	10.5
0.50	$2.5 \times 10^7$	$1.7 \times 10^6$	114	7.7
0.75	$1.9 \times 10^7$	$1.3 \times 10^6$	85	5.7
1.0	$1.4 \times 10^7$	$9.4 \times 10^5$	63	4.2
1.25	$1.1 \times 10^7$	$7.0 \times 10^5$	47	3.1
1.5	$7.8 \times 10^6$	$5.1 \times 10^5$	35	2.3
1.75	$5.8 \times 10^6$	$3.8 \times 10^5$	26	1.7
2.0	$4.2 \times 10^6$	$2.8 \times 10^5$	19	1.2
2.25	$3.2 \times 10^6$	$2.0 \times 10^5$	15	0.9
2.50	$2.4 \times 10^6$	$1.5 \times 10^5$	11	0.7
2.75	$1.8 \times 10^6$	$1.1 \times 10^5$	8	0.5
<u>3 thru 8</u>	<u>---</u>	<u>---</u>	<u>24</u>	<u>1.4</u>
		Total curies	600	39.9

TABLE 9

COBALT-60, NICKEL-63, AND NICKEL-59 IN THE DR THERMAL SHIELD  
MARCH 1, 1985

<u>Shield Section</u>	<u>Cobalt-60 Curies</u>	<u>Nickel-63 Curies</u>	<u>Nickel-59 Curies</u>
Right	600	40	0.32
Left	600	40	0.32
Top	600	40	0.32
Bottom	600	40	0.32
Front*	780	52	0.42
Rear*	<u>780</u>	<u>52</u>	<u>0.42</u>
TOTAL	3960	264	2.1

\*Corrected for volume and mass.

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The cobalt-60 and nickel-63 inventories for the reactor thermal shields were estimated by comparing the DR Reactor sample data (Appendix A) to the other reactors. Where sample data were not available, e.g., nickel-59, the inventories were calculated based on the ratio of nickel-59 to nickel-63 described in Appendix E. To determine the DR Reactor shield inventory, the sample data were plotted on graph paper to find the specific activity as a function of distance from the inner face of the shield. By considering the shield to be made up of a series of 1/4-in. thick slabs with the same height and length as the adjacent graphite stack, the total curie inventory for each slab could be estimated.

The cobalt-60 inventory estimate for the DR Reactor was adjusted for the different operating times and decay times for the other older reactors. The K Reactors were additionally adjusted for the greater mass of the shields. Nickel-63 and nickel-59 inventories were adjusted only for the different operating times and masses since the difference due to varying decay times is negligible.

The thermal shield cooling tubes contribute significantly to the nickel-63 and nickel-59 inventories since the stainless steel used for the tubes was 8 % nickel. The inventory estimates for the radionuclides in the cooling tubes were calculated using a flux level of  $10^{11}$  neutrons/cm<sup>2</sup>-s for the total equivalent operating days of each reactor. The K Reactors required a further adjustment to account for the increased number and length of the tubes.

Table 9 shows the estimated inventories for the reactor thermal shields and cooling tubes.

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TABLE 10

RADIONUCLIDE INVENTORIES FOR THE REACTOR THERMAL SHIELDS  
AS OF MARCH 1, 1985

<u>Reactor</u>	<u>Cobalt-60</u> <u>(Curies)</u>	<u>Nickel-63</u> <u>(Curies)</u>	<u>Nickel-59</u> <u>(Curies)</u>
B shield	8600	380	3
cooling tubes	90	460	4
C shield	9800	380	3
cooling tubes	90	460	4
D shield	7300	370	3
cooling tubes	80	440	4
DR shield	4000	260	2
cooling tubes	60	320	3
F shield	4800	310	2
cooling tubes	70	370	4
H shield	4200	300	2
cooling tubes	70	350	3
KE shield	17200	550	4
cooling tubes	260	650	5
KW shield	14300	510	4
cooling tubes	220	620	5

### 2.3.3 Process Tubes

Radionuclides associated with the process tubes are from two sources: activation of the tube materials and film deposits on the inner tube wall from corrosion products. Samples of three process tubes were taken from the DR Reactor for analysis (Appendix A). The analyses were performed on the process tube material and on the film layer. In order to apply the DR Reactor sample data to the entire reactor or to the other Hanford reactors, some clarification is necessary.

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The induced activity of the process tube material varies from front to rear of the reactor according to the neutron flux distribution. Ignoring local effects of control rods, the neutron flux distribution may be approximated by a cosine over the length of interest. That is, the flux level is a maximum at the center of the reactor and drops by several orders of magnitude several feet past the end of the fuel charge.

The activity of the film layer inside the process tubes starts at a low value near the front face end of the fuel charge (upstream end) and reaches a maximum value past the rear face end of the fuel charge (downstream end). Since the reactors in this document used "single pass" cooling in which cooling water was not recirculated, the activity along the process tube film maintained the "S" shape distribution throughout the life of the tube.

The DR Reactor process tube samples were obtained about one-fourth of the fuel charge length from the downstream end. At this position the neutron-induced activity in the process tube was lower than at the center of the tube but overall was nearly average for the entire length of tube. The activity of the film was less than the maximum concentration, but provides a conservative estimate when applied to the entire tube length, since film upstream of the sample point would be of lower activity.

The DR Reactor process tube film samples were apparently not analyzed for the nickel isotopes in this document. The fuel element cladding contained about 1% nickel. Some of the cladding material combined with the impurities in the process water and became part of the film on the process tubes. For the older reactors the nickel-63 inventory of the process tubes was estimated to be about 10 curies. If the nickel-63 is 10 curies, then the nickel-59 inventory is about 0.1 curies. Since a film analysis was not performed for the K Reactors, a factor of 1.5 was applied to the older reactor values to account for the greater number of process tubes and the greater length of the tubes.

A chemical analysis was made of a piece of unirradiated Zircaloy-2 process tube to identify elements that would produce long half-life nuclides by neutron activation. The elements of interest are nickel, zirconium, niobium, and molybdenum. The analysis results were stated earlier in Section 2.2.3. Calculations were made for the estimated inventories of zirconium-93, nickel-63, and nickel-59 in the Zircaloy-2 process tubes for the K Reactors. The estimated inventories for molybdenum-93 and niobium-94 were calculated from the data set given in Reference 1 with adjustment for the actual exposure time of the Zircaloy-2 process tubes in the K Reactors.

A summary of the estimated radionuclide inventory for the reactor process tubes is given in Table 11.

TABLE 11

ESTIMATED RADIONUCLIDE INVENTORY IN THE  
REACTOR PROCESS TUBES

Radio- Nuclide	Reactor (Curies)							
	<u>B</u>	<u>C</u>	<u>D</u>	<u>DR</u>	<u>F</u>	<u>H</u>	<u>KE</u>	<u>KW</u>
<sup>60</sup> Co	300	350	270	200	210	200	190	170
<sup>59</sup> Ni	0.1	0.1	0.1	0.1	0.1	0.1	13	11
<sup>63</sup> Ni	10	10	10	10	10	10	1700	1500
<sup>90</sup> Sr	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.3
<sup>93</sup> Mo	--	--	--	--	--	--	0.2	0.2
<sup>93</sup> Zr	--	--	--	--	--	--	11	10
<sup>152</sup> Eu	1.6	1.7	1.5	1.3	1.4	1.3	2	2
<sup>154</sup> Eu	1.2	1.3	1.1	0.9	1.0	1.0	1.6	1.6

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#### 2.3.4 Reactor Control System

The vertical safety rods, horizontal controls rods, and ball 3X safety system make up the reactor control system. Only a small segment of each control rod was exposed to the reactor's neutron fluence. Additionally, only the reactor entry ports of the ball system were exposed. The degree to which these exposed portions became radioactive depended on the component's chemical composition. The radionuclide inventories of the vertical and horizontal control rods are assumed to be equal to the amounts of thermal shield removed by the penetrations. See Section 2.2.4.

When the ball 3X system balls were dropped into the reactor, a small number of balls became trapped in the openings of the graphite and could not be removed. These trapped balls caused a reactivity loss which was overcome by adding more enriched fuel. The amount of enrichment needed was equated to the mass of the balls in the reactor. For DR Reactor, this equated to 72 pounds of balls. Measured data on the gamma emission of a small number of irradiated balls were used to compute the amount of cobalt-60 that would be present in the trapped balls. Using this method, a cobalt-60 inventory of 110 curies was computed for the balls remaining in the DR Reactor core. The DR Reactor had the greatest amount of enriched fuel added to compensate for the balls in the stack. As a conservative measure 110 curies of cobalt-60 were added to each of the other seven reactor inventories.

#### 2.3.5 Trace Radionuclides

Impurities in the materials exposed to the neutron flux became activated during reactor operation. While the contribution of these activated impurities is small compared to the major constituents considered above, calculations were performed to indicate the amounts of trace radionuclides that may be present in various reactor components. The calculations in most cases were based on the data set provided in Reference 1 with neutron flux levels of  $10^{14}$ ,  $10^{11}$ , and  $10^{10}$  assumed for the graphite stacks, the

thermal shields, and the biological shields respectively. In cases where an impurity level had not been chemically analyzed, the suggested value given in Reference 1 was used for the calculations. In the case of technicium-99 production from neutron activation of molybdenum-98, a manual calculation was made using basic activation formula. No corrections or adjustments were made to determine the trace radionuclides for each reactor. Instead, the inventories were grouped by the older reactors and the K reactors.

Tables 12 and 13 show the impurity level of the parent isotope, the radionuclide produced, and the calculated curie content of the reactor component.

TABLE 12

TRACE RADIONUCLIDE INVENTORIES FOR  
B, C, D, DR, F AND H REACTOR COMPONENTS

<u>Component</u>	<u>Parent Nuclide and Concentration (ppm)</u>		<u>Radionuclide Formed</u>	<u>Curies Per Component</u>
Graphite stack	Niobium	0.15	$^{94}\text{Nb}$	0.3
Thermal shield	Silver	0.35	$^{108}\text{Ag}$	0.03
	Niobium	4.9	$^{94}\text{Nb}$	0.02
	Molybdenum	210	$^{93}\text{Mo}$	0.04
			$^{99}\text{Tc}$	0.002
Concrete Base	Calcium	72000	$^{41}\text{Ca}$	2
Top biological shield (C Reactor only)	Calcium	72000	$^{41}\text{Ca}$	2

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TABLE 13

TRACE RADIONUCLIDE INVENTORIES FOR  
KE AND KW REACTOR COMPONENTS

<u>Component</u>	<u>Parent Nuclide and Concentration (ppm)</u>		<u>Radionuclide Formed</u>	<u>Curies Per Component</u>
Graphite stack	Niobium	0.15	$^{94}\text{Nb}$	1.1
Thermal shield	Silver	0.35	$^{108}\text{Ag}$	0.04
	Niobium	4.9	$^{94}\text{Nb}$	0.03
	Molybdenum	210	$^{93}\text{Mo}$	0.06
			$^{99}\text{Tc}$	0.003
Zircaloy-2 Process Tubes	Niobium	14	$^{94}\text{Nb}$	0.6
	Molybdenum	50	$^{93}\text{Mo}$ $^{99}\text{Tc}$	0.2 0.03
Concrete Base and Biological Shield	Calcium	72000	$^{41}\text{Ca}$	15

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### 3.0 FUEL STORAGE BASINS

Each reactor building contains an irradiated fuel storage basin. The basin served as a collection, storage, and transfer facility for the fuel elements discharged from the reactor. A typical reactor fuel storage basin consists of the fuel element pickup chute area, the storage area, and the transfer area. Irradiated fuel elements were sorted in the pickup chute area and hand-tonged into storage buckets, and then transferred by an overhead monorail system to the storage area where they were held for the decay of short-lived radionuclides. Following the storage period, the buckets of fuel elements were moved by the overhead monorail system to the transfer area, where they were placed in railroad cask cars for transport to the chemical reprocessing facilities in the Hanford 200 Area.

During the past two years work has been initiated to clean up and stabilize the basins prior to decommissioning. The D and DR basins were washed down, the sludge was removed and taken to the 200 Area waste disposal site, and an asphalt emulsion (ATCO 1840) was applied to the floor and walls to fix any remaining contamination. At the B and C Reactor basins, the sludge was moved to the lower part of the fuel transfer pits and shielded with a cap, and the walls above the sludge in the pits, and the walls and floors of the basins were coated with the asphalt emulsion. The K Reactor storage basins are still in operation, storing irradiated fuel from the N Reactor. The F and H Reactor fuel storage basins were backfilled with soil in 1970. The soil covers the sludge and miscellaneous equipment that were in the basins at the time.

### 3.1 METHODS AND SOURCES FOR FUEL STORAGE BASINS

The radionuclides in the fuel storage basins originated from process tube scale and from failed fuel elements that were discharged into the basins. Over the years sludge accumulated on the floors of the basins. The sludge is the primary source for the basin inventory.

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Sample data were obtained from the sludges prior to removal and estimates of residual inventories were made from survey data taken after cleanout. As the B Reactor basin had the highest overall inventory prior to cleanout, that inventory was used as an estimate for the F and H Reactor basins. The K Reactor basins will be cleaned prior to decommissioning, hence, the inventory of the DR Reactor basin is applied to the K Reactor basins.

The water was removed from the B and C fuel storage basins. The contaminated sludge was pumped into the transfer area pits. The basin walls were washed down using high pressure water and then coated with an asphalt-based emulsion to fix the residual contamination to the surface. Additional characterization of the sludge in the transfer area pits may be required prior to final disposition.

The status of the D and DR basins is the same as the B and C basins, except that the sludge was removed from the basins and buried in the 200 Area low-level waste burial ground.

KE and KW fuel storage basins are still in use to store N Reactor fuel. It is assumed that these basins will be drained and the sludge will be removed, packaged, and shipped for disposal. The basins' floors and walls will be cleaned and the contamination on the surfaces fixed.

After the shutdown of F and H Reactors, the irradiated fuel was shipped to the 200 Area, the storage basins' water was pumped down to within a few feet above the basin floor and a detailed radiological and visual survey was conducted of the basins. After the completion of the survey and removal of identified high-dose-rate materials, the two basins were backfilled with soil in 1970. Irradiated spacers, process tubing, buckets, and miscellaneous reactor hardware were not removed prior to backfilling. Additionally, there is a possibility that one or more low-dose-rate irradiated fuel elements

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were overlooked and buried under the 20 feet of earth backfill. If the soil fill is removed from the F and H basins, the fuel elements, if any, will be shipped to the 200 Areas for final disposition and the sludge mixed with the soil will be packaged and shipped to the 200 Area low-level waste burial site. If the decision is made not to remove the soil from the basins, the sludge inventory from the B Reactor basin (highest inventory of all the basins) and a conservative inventory of possible fuel elements should be added. The calculated inventory of five fuel elements is shown in Table 14.

TABLE 14

ESTIMATED INVENTORY OF FIVE SPENT FUEL  
ELEMENTS (Assumes 15-year-old Fuel)

<u>Radionuclide</u>	<u>Half-life (years)</u>	<u>Curies</u>
<sup>90</sup> Sr	29	40
<sup>137</sup> Cs	30.2	40
<sup>152</sup> Eu	13.4	0.0003
<sup>238</sup> Pu	87.7	0.02
<sup>239</sup> Pu	24110	1.2
<sup>240</sup> Pu	6537	0.3
<sup>241</sup> Pu	14.7	10
<sup>85</sup> Kr	10.7	2
<sup>151</sup> Sm	90	0.4
<sup>113</sup> Cd	14.6	0.002
<sup>94</sup> Nb	20000	0.0008
<sup>79</sup> Se	65000	0.0002
<sup>107</sup> Pd	6500000	0.00002
<sup>241</sup> Am	432	0.4
<sup>99</sup> Tc	213000	1
<sup>93</sup> Zr	1500000	0.002
<sup>U</sup> <sub>238</sub>	4500000000	0.006

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### 3.2 CALCULATION ADJUSTMENTS AND ASSUMPTIONS FOR FUEL STORAGE BASINS

It is estimated that each of the basins at B, C, F, and H Reactors contains 50,000 kg of sludge. The radionuclide inventories for the fuel storage basins that contain sludge were calculated by multiplying the 50,000 kg by measured nuclide concentrations. The inventories for D and DR Reactor basins were determined from surveys taken after cleanout of the basins. The KE and KW Reactor basins are still in use to store irradiated fuel from N Reactor. It is assumed that the K Reactor basins will be cleaned at least to the present levels at the D and DR basins prior to decommissioning.

During the cleanout of the D and DR Reactor basins five irradiated fuel elements (two at D and three at DR) were found in the sludge on the basin floors. The layaway procedures used at the D and DR Reactors were much the same as used at the F and H Reactors. Since fuel elements were found at D and DR, it must be assumed that elements could also be present in the F and H Reactor basins. To provide a basis for the EIS evaluation, the radionuclide inventory for five fuel elements are listed.

Should the decision be made not to remove the earthen fill from the basins to search for spent fuel, the estimated inventories for F and H should be adjusted to add the inventory of the radionuclides identified as part of the fuel elements and estimated sludge.

### 3.3 ESTIMATES FOR FUEL STORAGE BASINS

Based on the information stated, the fuel storage basin estimated inventories are listed in Table 15.

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TABLE 15.

FUEL STORAGE BASIN INVENTORY  
CURIES AS OF MARCH 1, 1985

Basin	$^{59}\text{Ni}$	$^{60}\text{Co}$	$^{63}\text{Ni}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{152}\text{Eu}$	$^{154}\text{Eu}$	$^{238}\text{U}$	$^{239}\text{Pu}$	$^{241}\text{Am}$	$^{238}\text{Pu}$
B	0.5	11	60	14	16	1.4	4.2	0.009	1.6	0.5	0.075
C	0.16	16	16	7	6	4	7	0.004	1.5	0.5	0.075
D	0.002	0.05	0.27	0.06	0.12	0.02	0.007	--	0.024	0.008	--
DR	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
F*	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
H*	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
KE	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
KW	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
TOTAL											

\*Assumes that F and H basins are cleaned to same level as the DR basin. If soil is left in the basins, assume B basin sludge inventory and add fuel elements (see text).

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#### 4.0 CONTAMINATED AND ACTIVATED PORTIONS OUTSIDE THE REACTOR BLOCK AND FUEL STORAGE BASIN

There are reactor components and numerous facilities that are radioactively contaminated but not included in the previously discussed inventories. These components or locations are estimated to contain less than 5% of the total reactor facility inventory; therefore, exact inventories were not estimated. As examples, the soil columns under a fuel storage basin and reactor may be contaminated. These areas have not been characterized, but the inventory is estimated to be low when compared to the total inventory for the reactor facility.

While not within the stated scope of this document, several ground disposal facilities in the near vicinity of the reactor buildings were examined. If the in situ option for decommissioning the reactor were chosen, five ground disposal facilities would be included in the 210-ft radius of the earthen mound. The affected facilities are the 116-B-3 Pluto Crib, the 116-B-4 Dummy Decontamination Drain, the 116-KE-1 115 Crib, the 116-KW-1 115 Crib, and the 105-C Ball 3X Silos. The calculated inventories for these facilities are included in Appendix F for information.

#### 4.1 METHOD AND SOURCES FOR PORTIONS OUTSIDE THE REACTOR BLOCK AND BASINS

Radiation surveys of reactor building areas other than the reactor blocks and fuel storage basins, indicated general radiation levels of less than 1 mrem/hr to 5 mrem/hr, with isolated areas reading up to 10 mrem/hr. The areas of the survey included the front and rear work areas of the reactor, the pipe tunnels, the outer rod room, and other miscellaneous access areas within the reactor buildings.

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#### 4.2 CALCULATION ADJUSTMENTS AND ASSUMPTIONS FOR PORTIONS OUTSIDE THE REACTOR BLOCK AND BASINS

There were no inventory estimates made of the radionuclides that may be present in the form of contamination in reactor building spaces outside the reactor blocks and fuel storage basins. As noted in Section 4.1, the dose rates measured during a radiological survey were quite small.

9 3 1 2 8 7 2 0 4 5 4

## 5.0 SUMMARY OF RADIONUCLIDE INVENTORIES

The inventory estimates presented in this document are summarized in Tables 16-23 for each of the surplus reactor facilities. The total radionuclide inventory for a reactor facility ranges from 13,000 curies to just over 58,000 curies for the two reactor types, as shown by the tables.

The quantities stated in the inventories are conservative, as the methodology was designed to overestimate the radionuclides in the facilities. As examples of the conservatism, sample points in the thermal shield and the process tubes were selected for higher than average concentrations for a component.

The thermal shield calculations are based on the samples measured toward the center of the right side shield. Even though the samples would represent a higher fluence area of the shield, therefore a higher concentration of radionuclides, the same concentration of radionuclides was assumed for the entire shield.

The same type of conservatism was included in the process tube estimates. Samples were taken in the central zone of the reactor and the measured radionuclide inventory was applied to the entire reactor without accounting for the much lower fluence in the tube ends and in the fringe regions of the reactor.

To be certain that some large source had not been overlooked and that the correct areas of the facilities were included, an independent technical review of the inventories was performed by persons knowledgeable in the field but not directly involved in the estimates. The suggestions and comments from the technical review were incorporated as appropriate into this final document.

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TABLE 16  
RADIONUCLIDE INVENTORY ESTIMATE  
B REACTOR SUMMARY  
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
<sup>3</sup> H	8300	--	--	--	--	--
<sup>14</sup> C	4500	--	--	--	--	--
<sup>41</sup> Ca	190	--	--	--	2	--
<sup>60</sup> Co	100	8690	300	110	--	11
<sup>59</sup> Ni	1	7	0.1	--	--	0.5
<sup>63</sup> Ni	180	840	10	--	--	60
<sup>36</sup> Cl	40	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.2	--	--	14
<sup>93</sup> Zr	--	--	--	--	--	--
<sup>93</sup> Mo	--	0.04	--	--	--	--
<sup>94</sup> Nb	0.3	0.02	--	--	--	--
<sup>99</sup> Tc	--	0.002	--	--	--	--
<sup>108</sup> Ag	--	0.03	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	16
<sup>152</sup> Eu	40	--	1.6	--	--	1.4
<sup>154</sup> Eu	20	--	1.2	--	--	4.2
<sup>238</sup> U	--	--	--	--	--	0.009
<sup>238</sup> Pu	--	--	--	--	--	0.075
<sup>239</sup> Pu	1	--	--	--	--	1.6
<sup>241</sup> Am	0.3	--	--	--	--	0.5

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TABLE 17  
RADIONUCLIDE INVENTORY ESTIMATE  
C REACTOR SUMMARY  
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio- Shield	Storage Basin
<sup>3</sup> H	8900	--	--	--	--	--
<sup>14</sup> C	4500	--	--	--	--	--
<sup>41</sup> Ca	14	--	--	--	4	--
<sup>60</sup> Co	60	9890	350	110	--	16
<sup>59</sup> Ni	--	7	0.1	--	--	0.16
<sup>63</sup> Ni	28	840	10	--	--	16
<sup>36</sup> Cl	85	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.2	--	--	7
<sup>93</sup> Zr	--	--	--	--	--	--
<sup>93</sup> Mo	--	0.04	--	--	--	--
<sup>94</sup> Nb	0.3	0.02	--	--	--	--
<sup>99</sup> Tc	--	0.002	--	--	--	--
<sup>108</sup> Ag	--	0.03	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	6
<sup>152</sup> Eu	40	--	1.7	--	--	4
<sup>154</sup> Eu	20	--	1.3	--	--	7
<sup>238</sup> U	--	--	--	--	--	0.004
<sup>238</sup> Pu	--	--	--	--	--	0.075
<sup>239</sup> Pu	1	--	--	--	--	1.5
<sup>241</sup> Am	0.3	--	--	--	--	0.5

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TABLE 18  
RADIONUCLIDE INVENTORY ESTIMATE  
D REACTOR SUMMARY  
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
<sup>3</sup> H	7700	--	--	--	--	--
<sup>14</sup> C	4300	--	--	--	--	--
<sup>41</sup> Ca	150	--	--	--	2	--
<sup>60</sup> Co	90	7380	270	110	--	0.05
<sup>59</sup> Ni	2	7	0.1	--	--	0.002
<sup>63</sup> Ni	280	810	10	--	--	0.27
<sup>36</sup> Cl	35	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.2	--	--	0.06
<sup>93</sup> Zr	--	--	--	--	--	--
<sup>93</sup> Mo	--	0.04	--	--	--	--
<sup>94</sup> Nb	0.3	0.02	--	--	--	--
<sup>99</sup> Tc	--	0.002	--	--	--	--
<sup>108</sup> Ag	--	0.03	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	0.12
<sup>152</sup> Eu	40	--	1.7	--	--	2
<sup>154</sup> Eu	20	--	1.2	--	--	0.007
<sup>238</sup> U	--	--	--	--	--	--
<sup>238</sup> Pu	--	--	--	--	--	--
<sup>239</sup> Pu	1	--	--	--	--	0.024
<sup>241</sup> Am	0.3	--	--	--	--	0.008

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TABLE 19

RADIONUCLIDE INVENTORY ESTIMATE  
DR REACTOR SUMMARY  
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio- Shield	Storage Basin
<sup>3</sup> H	4900	--	--	--	--	--
<sup>14</sup> C	3200	--	--	--	--	--
<sup>41</sup> Ca	90	--	--	--	2	--
<sup>60</sup> Co	30	4060	200	110	--	0.23
<sup>59</sup> Ni	1	5	0.1	--	--	0.01
<sup>63</sup> Ni	95	580	10	--	--	1.25
<sup>36</sup> Cl	55	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.2	--	--	0.29
<sup>93</sup> Zr	--	--	--	--	--	--
<sup>93</sup> Mo	--	0.04	--	--	--	--
<sup>94</sup> Nb	0.3	0.02	--	--	--	--
<sup>99</sup> Tc	--	0.002	--	--	--	--
<sup>108</sup> Ag	--	0.03	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	0.81
<sup>152</sup> Eu	40	--	1.3	--	--	0.23
<sup>154</sup> Eu	20	--	0.9	--	--	0.05
<sup>238</sup> U	--	--	--	--	--	--
<sup>238</sup> Pu	--	--	--	--	--	--
<sup>239</sup> Pu	1	--	--	--	--	0.024
<sup>241</sup> Am	0.3	--	--	--	--	0.008

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TABLE 20  
RADIONUCLIDE INVENTORY ESTIMATE  
F REACTOR SUMMARY  
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio- Shield	Storage Basin
<sup>3</sup> H	5800	--	--	--	--	--
<sup>14</sup> C	3700	--	--	--	--	--
<sup>41</sup> Ca	140	--	--	--	2	--
<sup>60</sup> Co	70	4870	210	110	--	0.23
<sup>59</sup> Ni	2	6	0.1	--	--	0.01
<sup>63</sup> Ni	190	680	10	--	--	1.25
<sup>36</sup> Cl	35	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.2	--	--	0.29
<sup>93</sup> Zr	--	--	--	--	--	--
<sup>93</sup> Mo	--	0.04	--	--	--	--
<sup>94</sup> Nb	0.3	0.02	--	--	--	--
<sup>99</sup> Tc	--	0.002	--	--	--	--
<sup>108</sup> Ag	--	0.03	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	0.81
<sup>152</sup> Eu	40	--	1.4	--	--	0.23
<sup>154</sup> Eu	20	--	1.0	--	--	0.05
<sup>238</sup> U	--	--	--	--	--	--
<sup>238</sup> Pu	--	--	--	--	--	--
<sup>239</sup> Pu	1	--	--	--	--	0.024
<sup>241</sup> Am	0.3	--	--	--	--	0.008

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TABLE 21

RADIONUCLIDE INVENTORY ESTIMATEH REACTOR SUMMARYAS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	<u>Graphite Stack</u>	<u>Thermal Shield</u>	<u>Process Tubes</u>	<u>Control System</u>	<u>Bio- Shield</u>	<u>Storage Basin</u>
<sup>3</sup> H	5500	--	--	--	--	--
<sup>14</sup> C	3500	--	--	--	--	--
<sup>41</sup> Ca	54	--	--	--	2	--
<sup>60</sup> Co	40	4270	200	110	--	0.23
<sup>59</sup> Ni	1	5	0.1	--	--	0.01
<sup>63</sup> Ni	120	650	10	--	--	1.25
<sup>36</sup> Cl	70	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.2	--	--	0.29
<sup>93</sup> Zr	--	--	--	--	--	--
<sup>93</sup> Mo	--	0.04	--	--	--	--
<sup>94</sup> Nb	0.3	0.02	--	--	--	--
<sup>99</sup> Tc	--	0.002	--	--	--	--
<sup>108</sup> Ag	--	0.03	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	0.81
<sup>152</sup> Eu	40	--	1.3	--	--	0.23
<sup>154</sup> Eu	20	--	1.0	--	--	0.05
<sup>238</sup> U	--	--	--	--	--	--
<sup>238</sup> Pu	--	--	--	--	--	--
<sup>239</sup> Pu	1	--	--	--	--	0.024
<sup>241</sup> Am	0.3	--	--	--	--	0.008

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TABLE 22

RADIONUCLIDE INVENTORY ESTIMATE  
KE REACTOR SUMMARY  
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio- Shield	Storage Basin
<sup>3</sup> H	30000	--	--	--	--	--
<sup>14</sup> C	7000	--	--	--	--	--
<sup>41</sup> Ca	1	--	--	--	15	--
<sup>60</sup> Co	5	17500	190	110	--	0.23
<sup>59</sup> Ni	--	9	13	--	--	0.01
<sup>63</sup> Ni	11	1200	1700	--	--	1.25
<sup>36</sup> Cl	300	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.3	--	--	0.29
<sup>93</sup> Zr	--	--	11	--	--	--
<sup>93</sup> Mo	--	0.06	0.2	--	--	--
<sup>94</sup> Nb	1.1	0.03	0.6	--	--	--
<sup>99</sup> Tc	--	0.003	0.03	--	--	--
<sup>108</sup> Ag	--	0.04	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	0.81
<sup>152</sup> Eu	40	--	2	--	--	0.23
<sup>154</sup> Eu	20	--	1.6	--	--	0.05
<sup>238</sup> U	--	--	--	--	--	--
<sup>238</sup> Pu	--	--	--	--	--	--
<sup>239</sup> Pu	1	--	--	--	--	0.024
<sup>241</sup> Am	0.3	--	--	--	--	0.008

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TABLE 23

RADIONUCLIDE INVENTORY ESTIMATEKW REACTOR SUMMARYAS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio- Shield	Storage Basin
<sup>3</sup> H	27000	--	--	--	--	--
<sup>14</sup> C	6700	--	--	--	--	--
<sup>41</sup> Ca	5	--	--	--	15	--
<sup>60</sup> Co	5	14500	170	110	--	0.23
<sup>59</sup> Ni	--	9	11	--	--	0.01
<sup>63</sup> Ni	15	1100	1500	--	--	1.25
<sup>36</sup> Cl	270	--	--	--	--	--
<sup>90</sup> Sr	10	--	0.3	--	--	0.29
<sup>93</sup> Zr	--	--	10	--	--	--
<sup>93</sup> Mo	--	0.06	0.2	--	--	--
<sup>94</sup> Nb	1.1	0.03	0.6	--	--	--
<sup>99</sup> Tc	--	0.003	0.03	--	--	--
<sup>108</sup> Ag	--	0.04	--	--	--	--
<sup>137</sup> Cs	30	--	--	--	--	0.81
<sup>152</sup> Eu	40	--	2	--	--	0.23
<sup>154</sup> Eu	20	--	1.6	--	--	0.05
<sup>238</sup> U	--	--	--	--	--	--
<sup>238</sup> Pu	--	--	--	--	--	--
<sup>239</sup> Pu	1	--	--	--	--	0.024
<sup>241</sup> Am	0.3	--	--	--	--	0.008

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## 6.0 HAZARDOUS MATERIALS

Hazardous materials were identified and inventoried on a recent inspection of the reactor facilities (Appendix G). As noted on the inventory sheets, much of the material could be removed without requiring either major dismantlement of equipment, walls or doors or special hoisting equipment other than a standard forklift. The disposition of hazardous waste and/or materials, including asbestos, mercury, polychlorinated biphenyl (PCB) oil, and other materials will be addressed in a Safety Hazards Assessment before actual decommissioning work begins on a facility. The applicable regulations will be reviewed and the Decommissioning Work Procedures will provide explicit instructions for disposal and control of hazardous materials during the decommissioning work. Should materials other than those shown in Appendix G be identified, they will be addressed at the time of definitive design. The materials identified in Appendix G and those listed below are contaminated or potentially contaminated with radioactive materials.

Not indicated in Appendix G is a significant quantity of lead contained within shielding doors. At each K Reactor facility, there are eight shield doors with an estimated inventory of about three tons of lead each. In addition, the thermal shield cooling tubes were embedded in the thermal shields with lead to provide a good thermal contact. Each of the older reactors has about 80 tons of lead associated with the cooling tubes. Each K Reactor has about 110 tons of lead associated with the cooling tubes.

Other sources of lead are the C Reactor Tool Dolly Room door (13.2 tons), the C Reactor Central Viewer Frame (4 tons), and the K Reactors Central Viewer Frames (1.3 tons each reactor).

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7.0 REFERENCE

1. L. D. Felstead and P. B. Woollam, An Assessment of all Known Isotopes to Determine Which Might be Important in the Decommissioning of Thermal Nuclear Reactors, TPRD/B/0386/N84, January 1984.

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APPENDIX A  
REACTOR CHARACTERIZATION

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APPENDIX ACHARACTERIZATION OF THE RADIOACTIVITY REMAINING IN THE  
SURPLUS PRODUCTION REACTORS AT HANFORDA.1 REACTOR SAMPLING

During 1976-1977, four sample holes were drilled into the DR Reactor block. (See Figure A-1.) The sample holes were drilled through the biological and thermal shielding and into the graphite stack. Sampling of the DR Reactor was intended to establish the following:

- o Radionuclide concentrations within the DR Reactor biological and thermal shields, and the graphite stack.
- o In-place reactor dose rates at selected locations.
- o Distribution of radionuclide concentrations in the thermal and biological shields with respect to depth of penetration.

In addition to the sampling and radiological monitoring performed at the DR Reactor, in-place dose rate measurements were taken of selected process tubes in the F and H Reactors.

Radioanalytical results of process tube sampling performed in 1967 at the DR Reactor are also reported along with decay corrections. In addition, samples had been taken of several gunbarrels from the C Reactor in 1967. The results of radionuclide analyses of these samples are also reported.

A.2 SUMMARY OF SAMPLING RESULTS

Reactor sampling at the experimental facility locations (X-levels) of the DR Reactor was initiated on December 28, 1976. Sampling of the first sample hole, A, was completed on January 28, 1977. Drill shaving samples were taken of the biological shield. A continuous core sample of the thermal shield was taken and segmented for radionuclide analyses. Two graphite samples were

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9 3 1 2 8 7 2 0 4 6 8

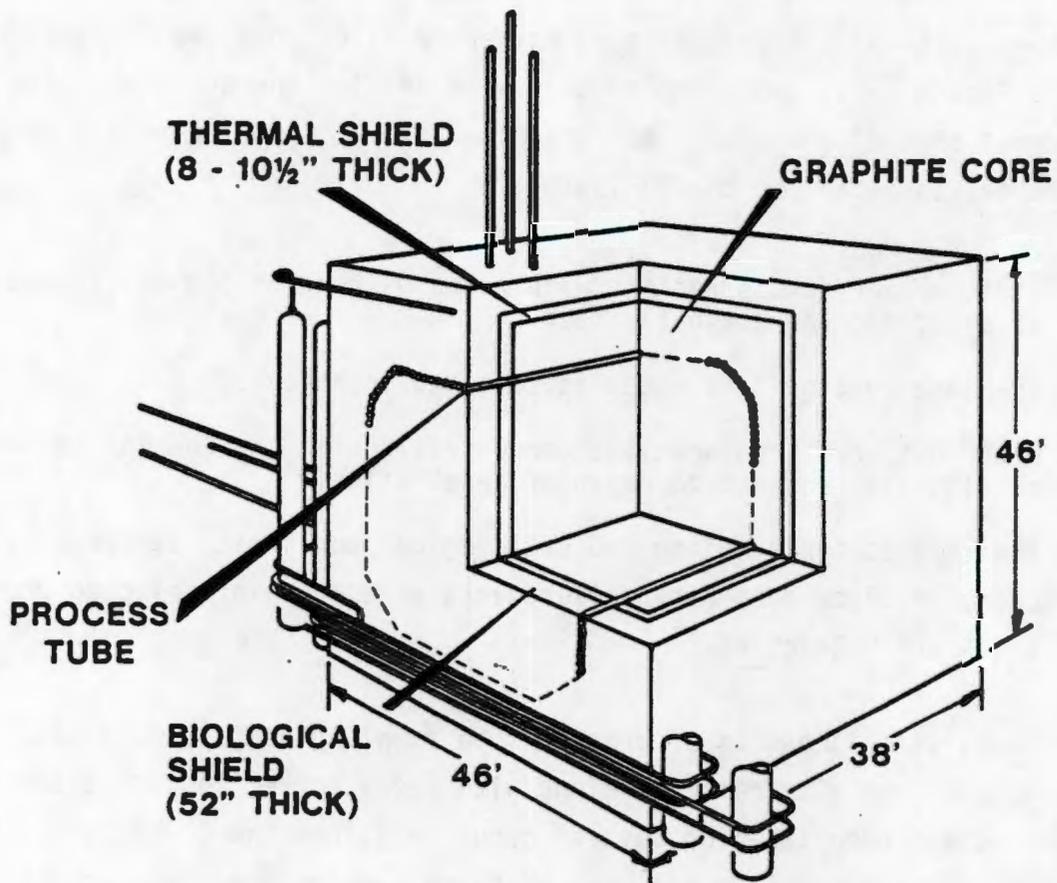
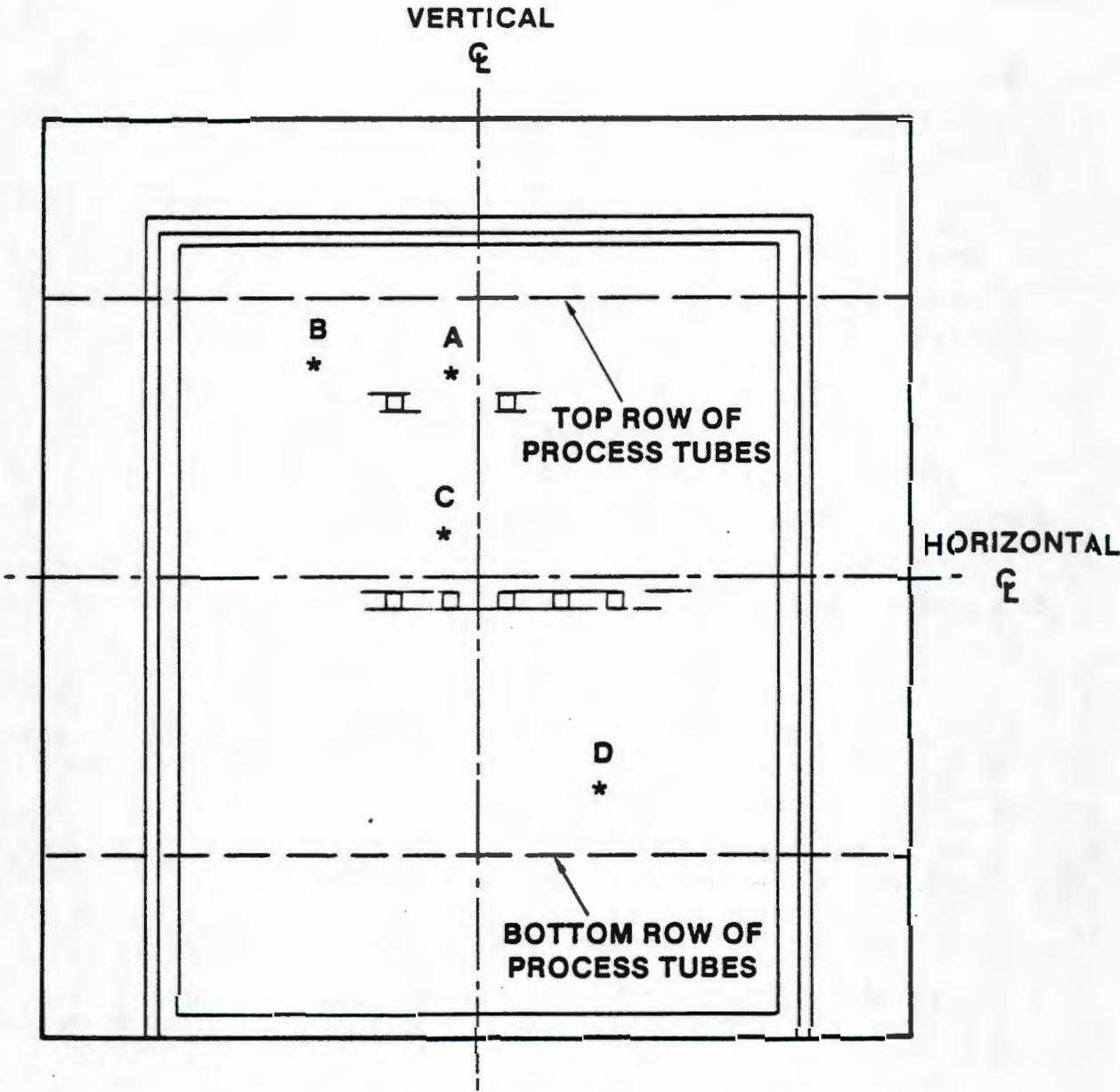


Figure A-1. Reactor Block Construction.



□ INDICATES EXISTING 4-3/16" SQUARE IRRADIATION TEST HOLES.

\* INDICATES SAMPLE HOLE

RIGHT SIDE VIEW

Figure A-2. Approximate Sample Hole Locations at the DR Reactor

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taken in the reflector area and five graphite samples were taken in the active portion of the reactor to a total distance of 33 feet, which is 6 feet from the reflector area at the opposite side of the reactor. The indicated dose rate on the 33-foot sample was 20 mR/hr. All other graphite samples were less than 2 mR/hr.

By March 14, 1977, a second sample hole, B, had been completed, and the biological and thermal shield samples of sample hole C had been taken. At this point, the sampling was temporarily terminated and the drilling equipment placed on standby until additional funding could be obtained to complete the work. Additional funding became available in late August 1977. The sampling work was restarted on August 29 and completed on September 22, 1977. Selected samples were submitted for radioisotopic analyses.

Some general conclusions with respect to the radioactivity in the DR Reactor are as follows:

- o In-place dose rates of selected process tubes average 50 R/hr in the vicinity of the gunbarrel tips and drop down to 4 R/hr 15 feet into the process tube from the nozzle.
- o In-place dose rates at the outer surface of the thermal shield were approximately 2 R/hr at the sample hole locations.
- o The maximum in-place dose rate measured was approximately 100 R/hr.
- o Approximately 90 percent of the biological shield radioactivity is contained in the inner 1-3/4-inch layer of steel closest to the thermal shield.
- o Approximately 90 percent of the total biological and thermal shield radioactivity is in the inner 2-inch portion of the thermal shield; 99 percent of the activity is in the inner 4-inch portion of the shield.
- o Approximately 98 percent of the thermal shield activity is from cobalt-60, and the remaining activity mostly from nickel-63. (Note: 1977 data)
- o Approximately 67 percent of the graphite activity is from tritium and 27 percent from carbon-14. (Note: 1977 data)
- o Plutonium-239/240 concentrations in the graphite stack average  $2.6 \times 10^3$  pCi/g up to a maximum of  $2.7 \times 10^4$  pCi/g.

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Some other general results reported in this study are as follows:

- o In-place dose rates of selected process tubes at the F and H Reactors average approximately 100 R/hr in the vicinity of the gunbarrel tips and drop down to 8 R/hr 15 feet into the process tubes from the nozzle.
- o Based on sampling performed in 1967, cobalt-60 concentrations in the DR Reactor process tubes are the order of  $2.5 \times 10^7$  pCi/g. Cobalt-60 concentrations in a mild steel gunbarrel tip removed from C Reactor are approximately  $1 \times 10^8$  pCi/g, and  $2 \times 10^9$  pCi/g in a stainless steel gunbarrel tip.

### A.3 PAST PROCESS TUBE AND GUNBARREL SAMPLING

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Sampling of the reactor hardware (process tubes, gunbarrels, etc.) was not performed as part of this program. Examples of typical dose rates of activated reactor hardware removed as part of maintenance projects during operating periods are: gunbarrels - 300 mR/hr to 4 R/hr at 3 feet; step-plugs - 100 mR/hr to 3 R/hr at 4 feet; horizontal control rods - 1 to 5 R/hr at 6 feet; and bundles of process tubing - 500 mR/hr to 5 R/hr at 6 feet.

A limited amount of sampling of the DR Reactor process tubes was performed in March 1967, a little more than 2 years after reactor shutdown. Sections of aluminum process tubes #3768, #1166, and #2568 were removed and analyzed for radionuclides. The film was removed from the interior of the tubes for separate analyses. All samples were taken 14 feet from the rear face van stone flange. The process tube connections are made to the gunbarrels by van stone flanges which were intended to prevent water from leaking into the stack and the gas atmosphere. A cross section of a process tube channel for DR Reactor is given in Figure A-3. The fuel element charges started approximately 8.5 feet from the end of the van stone flange.

Table A-1 lists the radioanalytical results for the process tube samples.

Table A-2 lists the same results as decay-corrected until March 1977, the midpoint of the reactor core sampling.

TABLE A-1  
RADIOANALYTICAL RESULTS OF PROCESS TUBE SAMPLES

<u>Radionuclide</u>	<u>Half-Life</u> (T 1/2)	<u>Process Tube Sections</u>		
		<u>pCi/g Aluminum</u>		
		<u>#3768</u>	<u>#1166</u>	<u>#2568</u>
<u>Tube</u>				
54Mn	314 day	2.4x10 <sup>7</sup>	2.4x10 <sup>7</sup>	3.8x10 <sup>8</sup>
60Co	5.3 year	9.4x10 <sup>7</sup>	9.2x10 <sup>7</sup>	4.6x10 <sup>8</sup>
<u>Film</u>				
54Mn	314 day	3.9x10 <sup>5</sup>	3.0x10 <sup>5</sup>	-
60Co	5.3 year	3.0x10 <sup>6</sup>	2.7x10 <sup>6</sup>	-
65Zn	245 day	8.8x10 <sup>6</sup>	5.1x10 <sup>6</sup>	-
152Eu	13 year	2.8x10 <sup>5</sup>	4.0x10 <sup>5</sup>	-
154Eu	8 year	5.0x10 <sup>5</sup>	2.8x10 <sup>5</sup>	-
90Sr	28 year	2.0x10 <sup>4</sup>		-

TABLE A-2  
RADIOANALYTICAL RESULTS OF PROCESS TUBE SAMPLES  
DECAY - CORRECTED TO MARCH 1977

<u>Radionuclide</u>	<u>No. of</u> <u>T 1/2's</u>	<u>Process Tube Sections</u>		
		<u>pCi/g Aluminum</u>		
		<u>#3768</u>	<u>#1166</u>	<u>#2568</u>
<u>Tube</u>				
54Mn	12	5.9x10 <sup>3</sup>	5.9x10 <sup>3</sup>	9.3x10 <sup>4</sup>
60Co	1.9	2.5x10 <sup>7</sup>	2.5x10 <sup>7</sup>	1.2x10 <sup>8</sup>
<u>Film</u>				
54Mn	12	9.5x10 <sup>1</sup>	7.3x10 <sup>1</sup>	
60Co	1.9	8.0x10 <sup>5</sup>	7.2x10 <sup>5</sup>	
65Zn	15	2.7x10 <sup>2</sup>	1.6x10 <sup>2</sup>	
152Eu	0.77	1.6x10 <sup>5</sup>	2.3x10 <sup>5</sup>	
154Eu	0.63	2.2x10 <sup>5</sup>	1.2x10 <sup>5</sup>	
90Sr	0.36	1.6x10 <sup>4</sup>		

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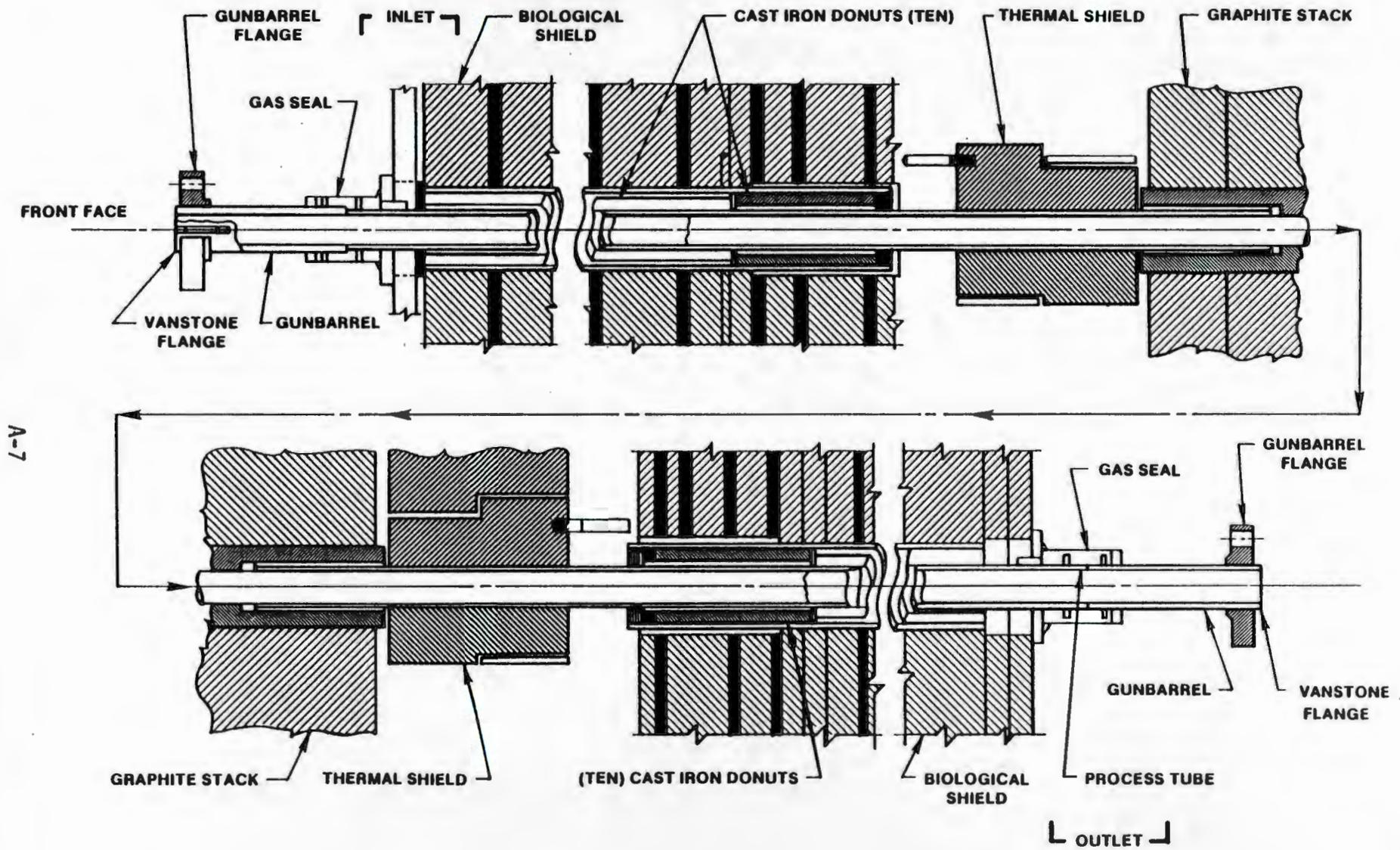


Figure A-3. Process Tube Channel Cross Section.

After 10 years of decay since the analyses were performed, cobalt-60 is the dominant radionuclide in the process tube wall. Cobalt-60, europium-152, europium-154, and strontium-90 are the primary radionuclides in the process tube film. Manganese-54 and zinc-65 have decayed to levels three orders of magnitude less than the other radionuclides. Manganese-54 and zinc-65 would probably be masked by the activity of the other radionuclides, if gamma analyses of process tube samples were now performed.

In April 1967 samples from the tips of two gunbarrels removed from C Reactor were analyzed. The cobalt-60 results for these two samples are given below.<sup>1</sup>

o C Reactor Gunbarrel (4475) - 304 stainless steel

$${}^{60}\text{Co} = 3.2 \times 10^9 \text{ pCi/g (Redox Lab Analysis)}$$

$${}^{60}\text{Co} = 7.9 \times 10^9 \text{ pCi/g (Purex Lab Analysis)}$$

o C Reactor Rear Gunbarrel (0954) - mild steel

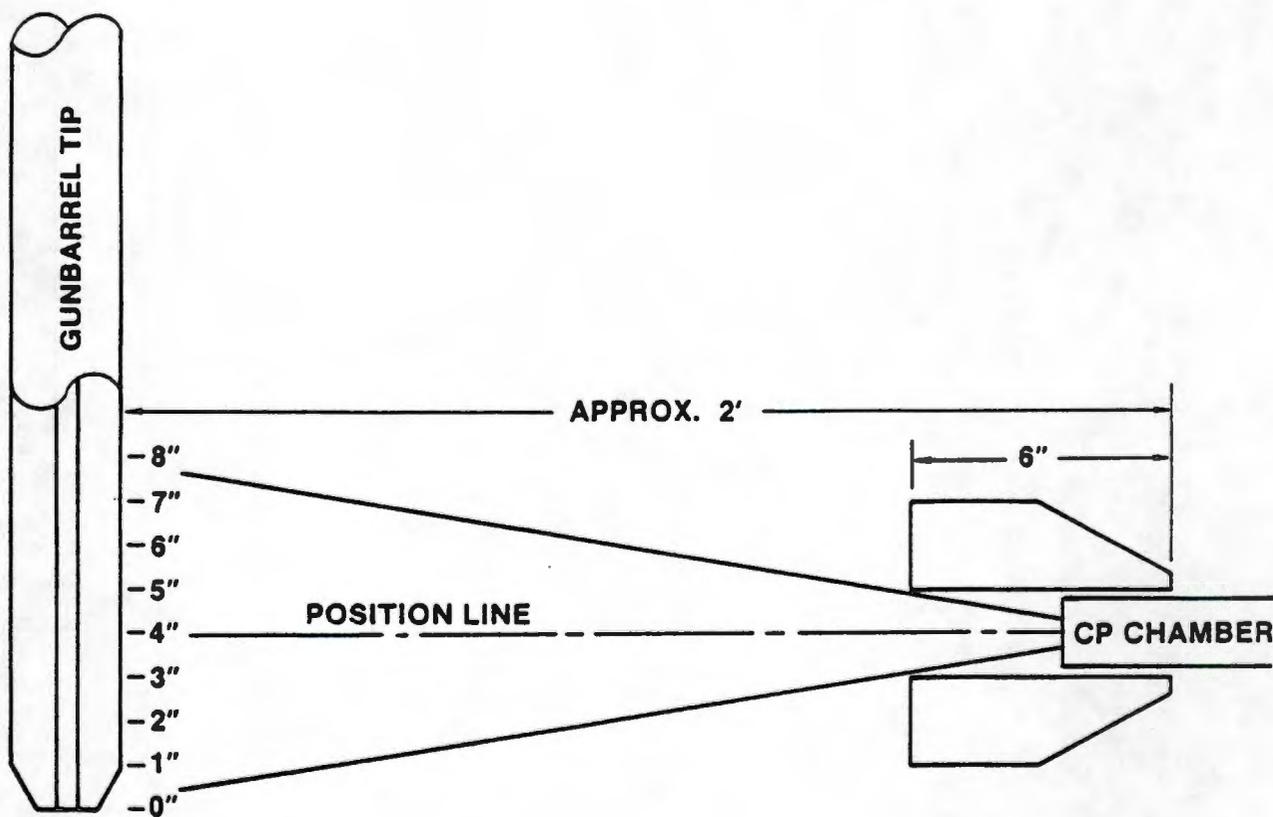
$${}^{60}\text{Co} = 4.4 \times 10^8 \text{ pCi/g (Redox Lab Analysis)}$$

Decay-correcting these data to March 1977 gives an average concentration of  $1.5 \times 10^9$  pCi/g for the stainless steel gunbarrel, and  $1.2 \times 10^8$  pCi/g for the mild steel gunbarrel. It should be noted that normal gunbarrels are mild steel. The stainless steel gunbarrel was one of about six prototypes installed on C Reactor for testing.

Dose rate measurements using a shielded VCP (with window closed) were taken of the mild steel gunbarrel to determine the extent of the gunbarrel tip activation. Figure A-4 illustrates the dose rate measurements taken along the tip of the gunbarrel. The dose rate information indicates that about a third of the gunbarrel was activated. (See Figure A-5.) The biological and thermal

<sup>1</sup>Other short half-life radionuclides (iron-59, cobalt-61, etc.) were detected, but would have since decayed away.

9 3 | 2 8 7 2 0 4 7 5



<u>POSITION (INCHES)</u>	<u>DOSE RATE (mR/hr)</u>
0	580
5	870
7	730
13	670
16	480
19	240
22	150
25	85

Figure A-4. Dose Rates to Determine Gunbarrel Tip Activation

A-10

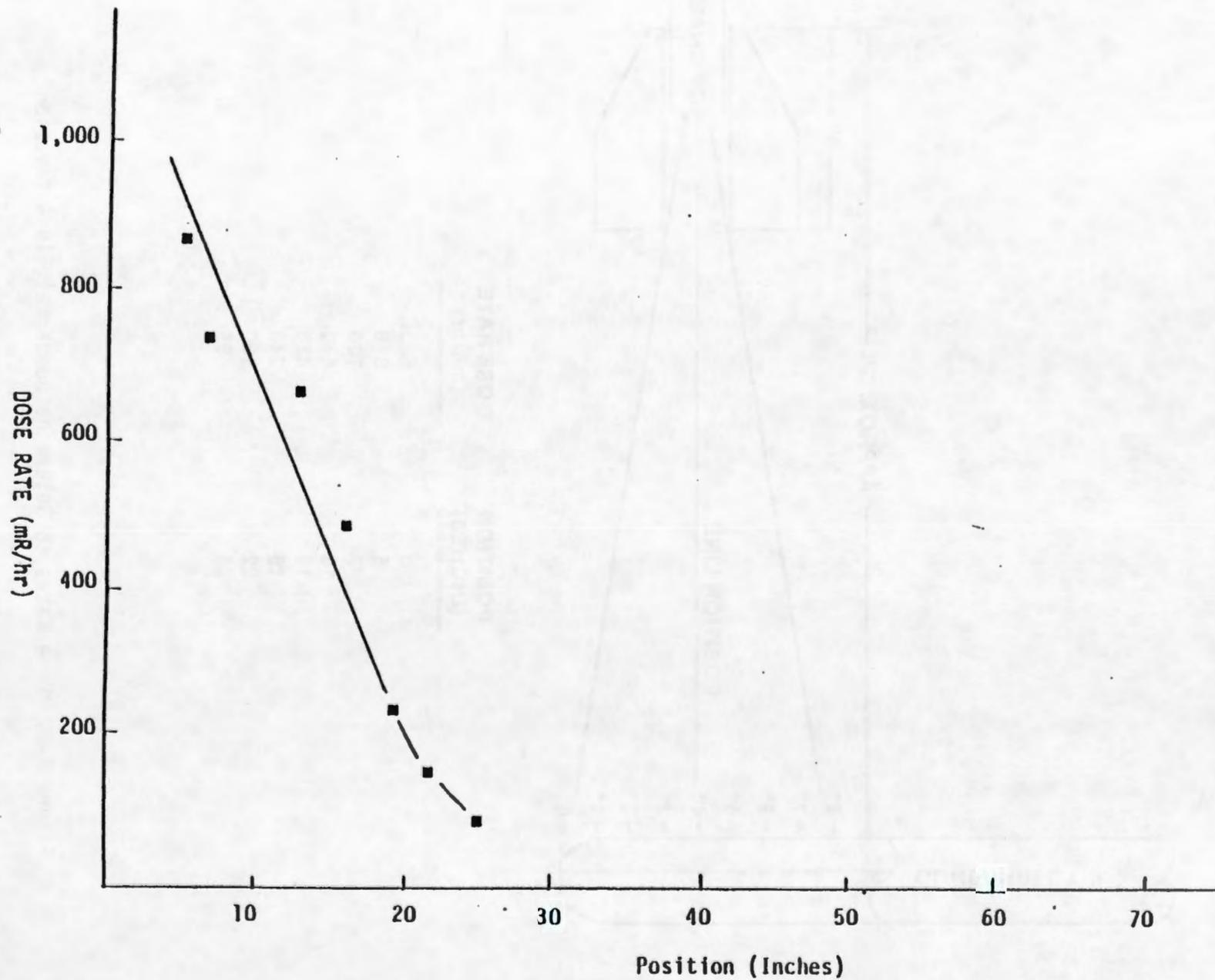


Figure A-5. C Reactor Gunbarrel Dose Rate Measurements at 2 Feet vs. Detector Position Line Along Gunbarrel Tip.

shield sampling performed as part of this study also indicated that the inner third of a gunbarrel will be activated.

#### A.4 DISCUSSION OF THE DR REACTOR CORE SAMPLING AND RESULTS

The approximate locations of the four sample holes drilled into DR Reactor from the X-levels were given in Figure A-2. A more precise description of the sample hole locations follows:

Sample Hole A - Located on the X-2 level of the right side of the reactor. The hole is in graphite layer number 80 from the bottom of the graphite stack and is approximately 10 feet above the reactor horizontal centerline and 5 inches left of the vertical centerline.<sup>2</sup>

Sample Hole B - Located on the X-2 level of the reactor. The hole is in graphite layer number 82 and is approximately 10 feet-8 inches above the reactor horizontal centerline and 7 feet-10 inches to the left of the vertical centerline.

Sample Hole C - Located on the X-1 level of the reactor. The hole is in graphite layer number 58 and is about 2 feet-3 inches above the reactor horizontal centerline and 1 foot-2 inches left of the vertical centerline.

Sample Hole D - Located on the X-0 level of the reactor. The hole is 2 feet-6 inches above the bottom row of process tubes and is between the 4th and 5th rows of process tubes. The hole is 13 feet below the reactor horizontal centerline and 7 feet right of the vertical centerline.

Basic sampling criteria were as follows:

- o Biological shield samples were drill cuttings of approximately 50 grams. The sampling interval of the biological shield samples depended upon the contamination levels encountered as measured by field instrumentation.
- o Thermal shield samples were 5/8-inch diameter core samples. Small aliquots were then taken for radioanalytical analyses.

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<sup>2</sup>There are 102 graphite layers in the DR Reactor graphite stack.

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- o Graphite stack samples were 1/2-inch diameter core samples taken approximately every 5 feet. The sample size was about 20 grams of graphite.

#### A.4.1 Biological Shield Samples

Reactor core samples were numbered for location and material type. 105-DR represents the DR Reactor building number.

- a) Masonite sample - 105-DR-AM1, where "A" designates the sample hole, and "M1" indicates that it was the first layer of masonite encountered in sample hole A. The second layer of masonite would be designated as 105-DR-AM2, etc.
- b) Iron sample - 105-DR-AI1, where "A" designates the sample hole, and "I1" indicates that it was the first layer of iron encountered in sample hole A.

#### A.4.2 Thermal Shield Core Samples

A thermal shield core sample is numbered 105-DR-AT, where "A" designates the sample hole and "T" that the sample is a thermal shield core sample.

Thermal shield samples were taken as continuous core samples, which were later segmented into smaller samples. Small aliquotes were taken as drill shavings collected at quarter-inch intervals along the core samples with the exception of test hole A. For test hole A, the thermal shield core sample was cut into quarter inch thick disks. Drill shavings were then taken by drilling through the face of the quarter inch thick disks. Drill shaving samples have suffixes of the form T1, T2, etc., where T1 indicates the inner quarter inch sample, T2 the following quarter inch sample, etc.

#### A.4.3 Graphite Samples

A reactor graphite sample is numbered 105-DR-AG15, where "A" designates the sample hole, and "G15" that it is a graphite sample taken from 15 feet into the reactor core.

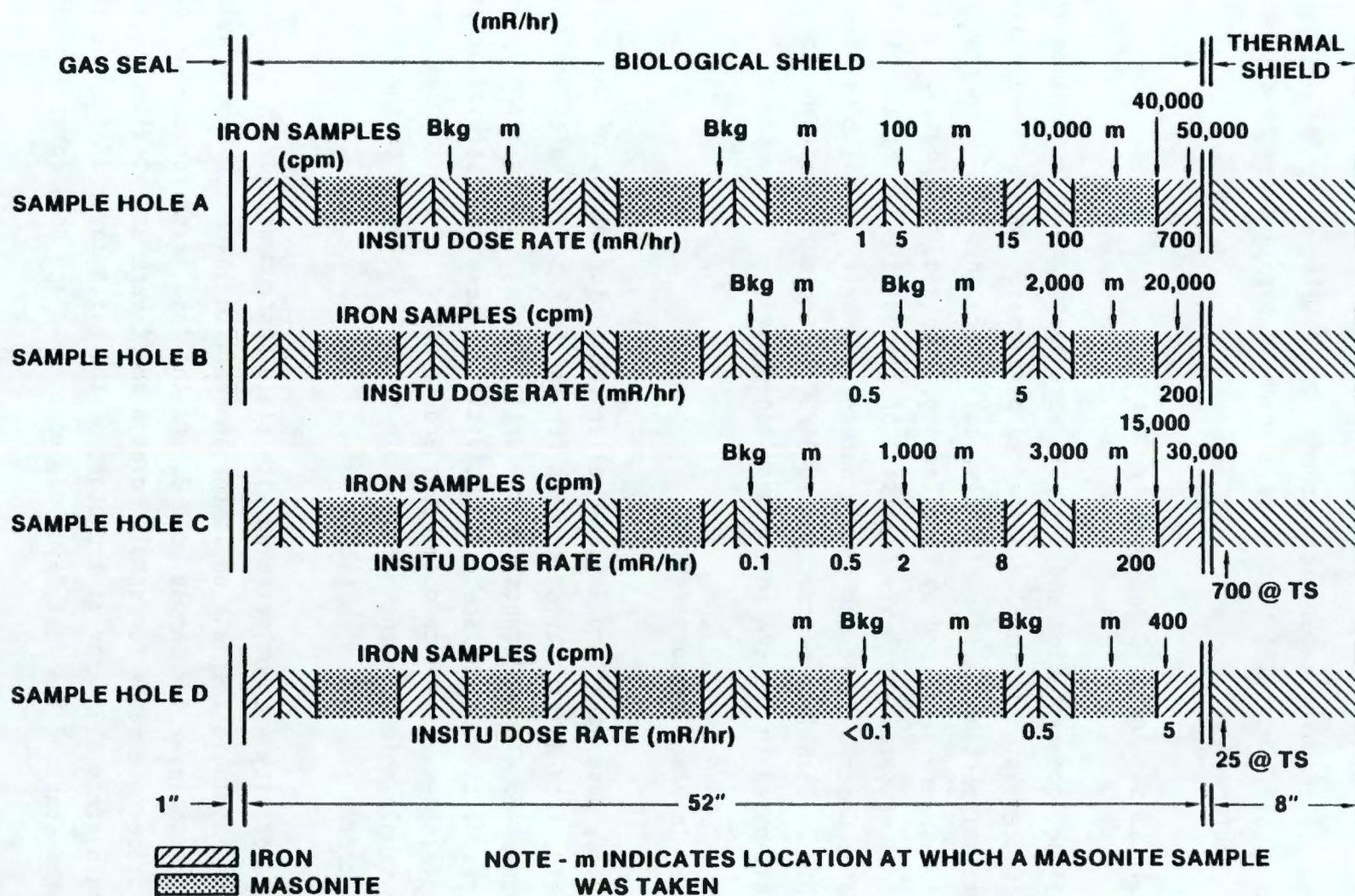
#### A.4.4 Sample Analysis and Quality Control

Sampling, documentation and analysis were performed in accordance with written procedures using trained, experienced personnel. Samples were analyzed for gamma emitters by UNC's radioanalytical laboratory. All other radionuclide analyses were performed by U.S. Testing, Richland, Washington. Quality control on sample analysis consisted of resubmitting a certain percentage of samples under a different sample number and verifying the consistency of results. Both labs had documented QA/QC controls in place and both participated in the EPA intercomparison program.

#### A.4.5 In-Place Measurements

In-place dose rates and Geiger-Mueller (GM) meter counts of the biological shield, iron-drill shaving samples for the four sample holes are shown in Figure A-6. The GM counts for the drill shaving samples are only partially representative of the specific activities because of possible variations in sample sizes. In-place dose rates and GM counts of the samples were highest for sample hole A, even though sample hole C is closest to the center of the reactor.

Radioactivity was not detected with field instrumentation in the iron samples of the biological shield until the inner fifth layer for sample holes A and C, the sixth layer for sample hole B, and the last layer for sample hole D. In-place dose rates for sample holes B and C were essentially the same, ranging from 0.5 mR/hr at the start of the fifth layer of iron up to 200 mR/hr at the start of the last layer of iron. In-place dose rates for sample hole A



A-14

Figure A-6. In-Place Dose Rates Within DR Reactor Sample Holes.

were the highest, ranging from 1 to 700 mR/hr for the same interval. Sample hole D had the lowest in-place dose rates which ranged from 0.1 mR/hr at the start of the fifth iron layer up to only 5 mR/hr at the start of the last layer of the biological shield.

No activity measurable with field instrumentation was detected in any of the Masonite samples, except for the last layer for sample hole A which read 1,000 cpm with a GM probe. This activity, however, is attributable to contamination from drill shavings which were visibly present in the sample. The contamination was removed from this Masonite sample by using a magnet.

Cobalt-60 concentrations for selected samples from the biological shields followed the same pattern indicated by field instrumentation, except that low concentrations of cobalt-60 were detected in the fourth layer of iron as shown in Table A-3.

TABLE A-3

COBALT-60 CONCENTRATION IN BIOLOGICAL SHIELD

Biological Shield Iron Layer	<sup>60</sup> Co Concentration (pCi/g)			
	Hole A	Hole B	Hole C	Hole D
7	$1.3 \times 10^5$	$5.4 \times 10^4$	$5.4 \times 10^4$	$4.0 \times 10^3$
6	$1.7 \times 10^4$	$6.8 \times 10^3$	$3.0 \times 10^3$	detect.
4	$3.3 \times 10^1$	$1.7 \times 10^1$	detect.	detect.

A comparison of thermal shield samples for these sample holes also shows the same trend (Table A-4). Sample holes B and C have similar concentrations, but hole A has concentrations about four times greater than B and C.

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TABLE A-4  
COBALT-60 CONCENTRATIONS IN THERMAL SHIELD

<u>Distance from Inner Surface of Thermal Shield</u>	<u><sup>60</sup>Co Concentration (pCi/g)</u>			
	<u>Hole A<sup>3</sup></u>	<u>Hole B</u>	<u>Hole C</u>	<u>Hole D</u>
1/4"	$4.0 \times 10^8$	$7.4 \times 10^7$	$7.6 \times 10^7$	$5.0 \times 10^7$
1/2"	$1.8 \times 10^8$	$4.8 \times 10^7$	$4.6 \times 10^7$	$3.5 \times 10^7$
1-1/2"	$2.6 \times 10^7$	$9.0 \times 10^6$	$1.8 \times 10^7$	$6.4 \times 10^6$
3"	$4.7 \times 10^6$	$1.4 \times 10^6$	$3.1 \times 10^6$	$9.6 \times 10^5$

More complete summaries of the radionuclide analyses of selected biological and thermal shield samples are included in Tables A-5 and A-6, respectively. The data indicate that approximately 90 percent of the total thermal and biological shield cobalt-60 activity is within the inner 1-3/4 inch of the thermal shield, and 99 percent of the activity is in the inner 4 inches of the thermal shield. The cobalt-60 activity within the inner half of the thermal shield has an average concentration of  $3.6 \times 10^7$  pCi/g.

Nickel-63 is also present in the thermal and biological shields. Because of the high expense of performing nickel-63 analyses, not all samples were analyzed for nickel-63. The highest nickel-63 concentration found within the inner 4 inches of the thermal shield was  $7.5 \times 10^6$  pCi/g.

Graphite samples had direct GM counts generally about 10,000 cpm, but ranged from 1,000 cpm to over 80,000 cpm. Uncorrected dose rate measurements of the samples with a Cutie Pie (CP)<sup>4</sup> were generally 1 mR/hr up to a maximum of 20 mR/hr for sample A-G33.

<sup>3</sup>Sample hole A results are adjusted for the difference in the technique used for segmenting the thermal shield core sample for this test hole; i.e., the concentration at a quarter inch is the average of samples T-1 and T-2, etc.

<sup>4</sup>Cutie Pie is the name given a hand-held, ionization-chamber-type, radiation detection instrument.

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TABLE A-5

DR REACTOR BIOLOGICAL SHIELD IRON SAMPLES

<u>Sample No.</u>	<u>Concentration (pCi/g)</u>	
	<u><math>^{60}\text{Co}</math></u>	<u><math>^{63}\text{Ni}</math></u>
A-I2	$2.6 \times 10^{-1}$	
I4	$3.3 \times 10^1$	
I5	$3.7 \times 10^2$	$1.2 \times 10^1$
I6	$1.7 \times 10^4$	
I7	$1.3 \times 10^5$	$3.4 \times 10^3$
I7-1/2	$9.7 \times 10^4$	
(3/4" core) I8	$2.5 \times 10^5$	
B-I4	$1.7 \times 10^1$	$5.7 \times 10^{-1}$
I6	$6.8 \times 10^3$	
I7	$5.4 \times 10^4$	$8.2 \times 10^2$
C-I4	*	
I5	$2.5 \times 10^3$	$4.1 \times 10^1$
I6	$3.0 \times 10^3$	
I7	$5.4 \times 10^4$	
I7-1/2	$6.8 \times 10^4$	$7.8 \times 10^2$
D-I4,5,&6	*	
I7	$4.0 \times 10^3$	$6.6 \times 10^1$

\*Less than analytical detection limit. Detection limit value data no longer available.

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TABLE A-6  
DR REACTOR THERMAL SHIELD SAMPLES

Concentration (pCi/g)					
Sample No.*	$^{60}\text{Co}$	$^{63}\text{Ni}$	Sample No.	$^{60}\text{Co}$	$^{63}\text{Ni}$
A-T1	$5.4 \times 10^8$	$4.8 \times 10^6$	C-T1	$7.6 \times 10^7$	$7.5 \times 10^6$
T2	$2.6 \times 10^8$		(1/2") T2	$4.6 \times 10^7$	
T3	$9.1 \times 10^7$		(1-1/2") T6	$1.8 \times 10^7$	
(1") T4	$7.8 \times 10^7$		(3") T12	$3.1 \times 10^6$	
T5	$4.6 \times 10^7$		T18	$8.2 \times 10^5$	
T6	$2.4 \times 10^7$		(6") T24	$2.5 \times 10^5$	$6.9 \times 10^3$
T7	$2.7 \times 10^7$		(7-1/4") T26	$1.4 \times 10^4$	
(2") T8	$2.5 \times 10^7$		D-T1**	$5.0 \times 10^7$	$9.2 \times 10^5$
T9	$2.7 \times 10^7$		T2	$3.5 \times 10^7$	
T10	$1.4 \times 10^7$		T3	$2.1 \times 10^7$	
(3") T11	$8.8 \times 10^6$		(1") T4	$1.2 \times 10^7$	
T12	$5.2 \times 10^6$		T5	$9.2 \times 10^6$	
T13	$4.1 \times 10^6$		T6	$6.4 \times 10^6$	
T14	$8.6 \times 10^6$		T7	$4.2 \times 10^6$	
(4") T15	$2.5 \times 10^6$		(2") T8	$4.2 \times 10^6$	
T16	$2.6 \times 10^6$	$3.9 \times 10^4$	(3") T9	$9.6 \times 10^5$	
T17	$1.6 \times 10^6$		(4") T10	$2.4 \times 10^5$	
B-T1	$7.4 \times 10^7$	$2.9 \times 10^6$	(5") T11	$7.8 \times 10^4$	$1.3 \times 10^3$
T2	$4.8 \times 10^7$				
T6	$9.0 \times 10^6$				
(3") T12	$1.4 \times 10^6$				
T18	$3.2 \times 10^5$				
(5") T20	$2.3 \times 10^5$	$5.0 \times 10^3$			

\*Thermal shield samples were taken as continuous 5/8-inch diameter cores. Drill shaving samples were taken at quarter-inch intervals and designated as T-1, T-2, etc.

\*\*For sample hole D, drill shaving samples were taken at 1/4-inch intervals for the first 2 inches, and then at 1-inch intervals.

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Radionuclide analyses for selected graphite samples from the DR Reactor are given in Table A-7. Tritium and carbon-14 are present in addition to mixed fission and activation products. Transuranics are also present.

Plutonium-239/240 concentrations average  $2.6 \times 10^3$  pCi/g. Sample A-G33 had a plutonium-239/240 concentration of  $2.7 \times 10^4$  pCi/g. Nickel-63 was present with concentrations of up to  $1.5 \times 10^5$  pCi/g.

At DR Reactor, there was enough stack separation and graphite breakage so that balls were known to have been retained in the stack structure after spurious ball-drops. In fact, one ball was recovered in a graphite sample taken from sample hole A drilled into the DR Reactor stack. The ball had uncorrected CP readings of 10 R/hr at one inch and 150 mR/hr at one foot. This undoubtedly was the case to various degrees at the other reactors as well. The ball 3X systems of the 100 Area production reactors utilized both 3/8-inch and 7/16-inch diameter nickel-plated boron steel, nickel-plated carbon steel, and stainless steel balls which dropped into the vertical safety rod channels after a trip of the ball 3X circuit. The nickel plating and nickel impurities in the stainless steel would be sources of nickel-63 radioactivity. Balls retained in the graphite stack would have been subjected continuously during reactor operation to the neutron flux, heat, and moisture within the stack.

All analytical results are accurate to two standard deviations. Additionally, for gamma-emitting radionuclides, the error is no greater than 30 percent of the result. Results with a greater error were recounted until the error was within the 30 percent limit.

#### A.5 REACTOR IN-PLACE DOSE RATE MEASUREMENTS

During February 1978 in-place dose rate measurements were taken in selected process tubes at the DR and F Reactors. The in-place dose rate measurements were taken using a Tele-probe. Similar measurements were taken at the H Reactor in August 1975. Readings were taken from both the front and rear

TABLE A-7

DR REACTOR GRAPHITE SAMPLES

Sample Number	Concentration (pCi/g)													
	Pu-238	Pu-239/ 240	Sr-90	U	<sup>3</sup> H	Eu-152	Co-60	Eu-154	Cs-134	Cs-137	Eu-155	C-14	Ba-133	Ni-63
A-G 6						5.6x10 <sup>4</sup>	8.0x10 <sup>4</sup>	4.6x10 <sup>4</sup>	1.0x10 <sup>3</sup>	2.0x10 <sup>3</sup>	1.2x10 <sup>4</sup>	5.4x10 <sup>5</sup>	4.9x10 <sup>3</sup>	1.2x10 <sup>5</sup>
G 7						4.3x10 <sup>4</sup>	6.1x10 <sup>4</sup>	1.7x10 <sup>4</sup>	3.2x10 <sup>3</sup>	1.1x10 <sup>3</sup>	4.5x10 <sup>3</sup>		3.5x10 <sup>3</sup>	
G 13	1.3x10 <sup>2</sup>	2.0x10 <sup>2</sup>	3.5x10 <sup>3</sup>	6.4x10 <sup>-1</sup>	9.8x10 <sup>6</sup>	7.7x10 <sup>4</sup>	1.1x10 <sup>5</sup>	1.8x10 <sup>3</sup>	1.1x10 <sup>3</sup>	7.1x10 <sup>3</sup>	1.1x10 <sup>3</sup>	2.7x10 <sup>6</sup>	7.4x10 <sup>3</sup>	
G 18	<1.4x10 <sup>2</sup>	3.8x10 <sup>1</sup>	1.7x10 <sup>3</sup>	2.0x10 <sup>0</sup>	1.3x10 <sup>7</sup>	1.9x10 <sup>3</sup>	2.7x10 <sup>3</sup>	9.9x10 <sup>1</sup>	1.2x10 <sup>2</sup>	3.0x10 <sup>3</sup>	1.3x10 <sup>2</sup>	7.1x10 <sup>6</sup>	3.4x10 <sup>3</sup>	
G 23						3.4x10 <sup>4</sup>	4.8x10 <sup>4</sup>	7.8x10 <sup>3</sup>	2.5x10 <sup>3</sup>	2.1x10 <sup>3</sup>	2.2x10 <sup>3</sup>	1.2x10 <sup>7</sup>	2.4x10 <sup>3</sup>	
G 28	<7.8x10 <sup>1</sup>	3.9x10 <sup>2</sup>	5.6x10 <sup>3</sup>	<1.1x10 <sup>0</sup>	7.1x10 <sup>6</sup>	1.3x10 <sup>5</sup>	1.8x10 <sup>5</sup>	6.6x10 <sup>3</sup>	1.8x10 <sup>3</sup>	1.6x10 <sup>4</sup>	1.9x10 <sup>3</sup>	4.3x10 <sup>6</sup>	1.4x10 <sup>4</sup>	
G 33*	4.7x10 <sup>4</sup>	2.7x10 <sup>4</sup>	7.5x10 <sup>6</sup>	8.1x10 <sup>-2</sup>	6.3x10 <sup>6</sup>	1.1x10 <sup>4</sup>	1.2x10 <sup>5</sup>	2.1x10 <sup>4</sup>	2.2x10 <sup>4</sup>	5.7x10 <sup>5</sup>	5.8x10 <sup>3</sup>	5.0x10 <sup>6</sup>	3.4x10 <sup>4</sup>	
B-G 6						5.7x10 <sup>2</sup>	8.2x10 <sup>2</sup>	1.4x10 <sup>2</sup>	2.1x10 <sup>1</sup>	3.5x10 <sup>2</sup>	3.8x10 <sup>1</sup>	8.2x10 <sup>5</sup>	2.8x10 <sup>2</sup>	
G 13	7.8x10 <sup>0</sup>	2.7x10 <sup>1</sup>	1.0x10 <sup>3</sup>	1.9x10 <sup>0</sup>	4.7x10 <sup>6</sup>	4.8x10 <sup>2</sup>	6.9x10 <sup>2</sup>	1.2x10 <sup>2</sup>	3.3x10 <sup>1</sup>	1.6x10 <sup>2</sup>	3.2x10 <sup>1</sup>	1.8x10 <sup>6</sup>	1.3x10 <sup>2</sup>	
G 18						6.2x10 <sup>3</sup>	8.9x10 <sup>4</sup>	1.0x10 <sup>4</sup>	1.4x10 <sup>3</sup>	1.3x10 <sup>4</sup>	2.7x10 <sup>3</sup>		1.0x10 <sup>4</sup>	
G 23	7.7x10 <sup>1</sup>	1.6x10 <sup>2</sup>	2.8x10 <sup>3</sup>	<6.2x10 <sup>-2</sup>	7.8x10 <sup>6</sup>	1.3x10 <sup>5</sup>	1.8x10 <sup>5</sup>	3.8x10 <sup>4</sup>	3.2x10 <sup>3</sup>	1.1x10 <sup>4</sup>	1.1x10 <sup>4</sup>	2.2x10 <sup>6</sup>	1.0x10 <sup>4</sup>	
G 28						2.1x10 <sup>4</sup>	3.0x10 <sup>4</sup>	5.9x10 <sup>3</sup>	2.6x10 <sup>3</sup>	1.1x10 <sup>4</sup>	1.1x10 <sup>3</sup>		4.4x10 <sup>3</sup>	
G 33	2.0x10 <sup>2</sup>	4.9x10 <sup>2</sup>	3.8x10 <sup>3</sup>		3.4x10 <sup>6</sup>	3.6x10 <sup>4</sup>	5.1x10 <sup>4</sup>	5.1x10 <sup>3</sup>	1.7x10 <sup>3</sup>	9.3x10 <sup>3</sup>	1.3x10 <sup>3</sup>	1.8x10 <sup>6</sup>	2.6x10 <sup>3</sup>	
C-G 7	2.9x10 <sup>1</sup>	<1.2x10 <sup>-1</sup>	1.0x10 <sup>3</sup>	<1.3x10 <sup>-1</sup>	2.4x10 <sup>6</sup>	1.2x10 <sup>5</sup>	4.3x10 <sup>4</sup>	1.4x10 <sup>5</sup>	9.7x10 <sup>2</sup>	1.6x10 <sup>3</sup>	2.9x10 <sup>4</sup>	3.8x10 <sup>5</sup>	5.5x10 <sup>3</sup>	5.1x10 <sup>4</sup>
G 13						3.3x10 <sup>2</sup>	8.9x10 <sup>2</sup>	3.4x10 <sup>2</sup>	2.0x10 <sup>1</sup>	4.5x10 <sup>1</sup>	7.6x10 <sup>1</sup>	2.8x10 <sup>6</sup>	7.7x10 <sup>1</sup>	
G 18	<1.0x10 <sup>1</sup>	<9.0x10 <sup>0</sup>	1.4x10 <sup>3</sup>	<1.0x10 <sup>0</sup>	4.1x10 <sup>6</sup>	4.9x10 <sup>1</sup>	1.3x10 <sup>2</sup>	5.0x10 <sup>1</sup>	2.9x10 <sup>0</sup>	1.3x10 <sup>3</sup>	1.1x10 <sup>1</sup>	2.2x10 <sup>6</sup>	2.2x10 <sup>3</sup>	
G 23	4.9x10 <sup>3</sup>	5.1x10 <sup>3</sup>	5.0x10 <sup>4</sup>	<2.5x10 <sup>0</sup>	5.0x10 <sup>6</sup>	1.9x10 <sup>3</sup>	8.5x10 <sup>3</sup>	3.3x10 <sup>3</sup>	1.7x10 <sup>3</sup>	6.5x10 <sup>4</sup>	7.2x10 <sup>2</sup>	3.3x10 <sup>6</sup>	1.1x10 <sup>5</sup>	1.5x10 <sup>4</sup>
G 28						7.9x10 <sup>2</sup>	2.1x10 <sup>3</sup>	8.1x10 <sup>2</sup>	4.7x10 <sup>1</sup>	2.8x10 <sup>3</sup>	1.8x10 <sup>2</sup>	1.5x10 <sup>6</sup>	4.8x10 <sup>3</sup>	
G 33						1.4x10 <sup>4</sup>	3.1x10 <sup>5</sup>	1.6x10 <sup>4</sup>	5.4x10 <sup>3</sup>	2.4x10 <sup>4</sup>	4.2x10 <sup>3</sup>	1.5x10 <sup>6</sup>	4.8x10 <sup>4</sup>	
G 38	2.5x10 <sup>2</sup>	2.0x10 <sup>2</sup>	5.3x10 <sup>3</sup>	<1.8x10 <sup>0</sup>	4.2x10 <sup>6</sup>	9.4x10 <sup>4</sup>	2.5x10 <sup>5</sup>	7.4x10 <sup>4</sup>	5.6x10 <sup>3</sup>	1.5x10 <sup>4</sup>	1.8x10 <sup>4</sup>	1.6x10 <sup>6</sup>	1.9x10 <sup>4</sup>	
D-G 7						2.4x10 <sup>5</sup>	4.5x10 <sup>4</sup>	2.8x10 <sup>5</sup>	7.1x10 <sup>2</sup>	3.0x10 <sup>3</sup>	4.5x10 <sup>4</sup>	6.1x10 <sup>5</sup>	8.2x10 <sup>3</sup>	
G 13	7.0x10 <sup>1</sup>	1.8x10 <sup>2</sup>	2.6x10 <sup>3</sup>	<2.6x10 <sup>-1</sup>	1.3x10 <sup>7</sup>	4.4x10 <sup>4</sup>	8.6x10 <sup>4</sup>	4.5x10 <sup>4</sup>	1.4x10 <sup>3</sup>	4.7x10 <sup>3</sup>	1.1x10 <sup>4</sup>	1.6x10 <sup>6</sup>	1.7x10 <sup>4</sup>	
G 18						1.6x10 <sup>4</sup>	6.0x10 <sup>4</sup>	1.9x10 <sup>4</sup>	2.6x10 <sup>3</sup>	1.0x10 <sup>4</sup>	4.0x10 <sup>3</sup>	2.7x10 <sup>6</sup>	1.3x10 <sup>3</sup>	
G 23	2.7x10 <sup>2</sup>	2.4x10 <sup>2</sup>	2.7x10 <sup>3</sup>	<4.0x10 <sup>0</sup>	7.7x10 <sup>6</sup>	1.0x10 <sup>5</sup>	2.0x10 <sup>5</sup>	1.0x10 <sup>5</sup>	3.2x10 <sup>3</sup>	1.3x10 <sup>4</sup>	2.4x10 <sup>4</sup>	2.0x10 <sup>6</sup>	1.7x10 <sup>4</sup>	1.5x10 <sup>3</sup>
G 28						1.8x10 <sup>5</sup>	1.8x10 <sup>5</sup>	1.8x10 <sup>5</sup>	1.2x10 <sup>3</sup>	1.0x10 <sup>4</sup>	7.4x10 <sup>3</sup>		1.9x10 <sup>4</sup>	
G 33						1.1x10 <sup>5</sup>	1.1x10 <sup>5</sup>	8.8x10 <sup>4</sup>	1.7x10 <sup>3</sup>	5.7x10 <sup>3</sup>	1.8x10 <sup>4</sup>		1.4x10 <sup>4</sup>	

\*Am-241 3.8x10<sup>3</sup>

Blanks in the table indicate that no analysis for that radionuclide was performed on a particular sample.

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faces up to 15 feet into the process tube from the end of the nozzle. Fifteen feet was the furthest distance the Tele-probe could be extended. The results of these readings are given in Table A-8. Two measurements are generally reported for each of the process tubes measured. The first reading is the maximum dose rate detected at the end of the gunbarrel, or about 9.5 feet from the nozzle. In two cases, three readings are reported. In these instances, in-place dose rate readings went down going away from the gunbarrel tip and then went back up, thereby indicating higher than normal activity in the graphite stack.

As part of the 100-F Area Decontamination & Decommissioning (D&D) program, additional in-place dose rate surveys of process tubes were initiated and completed on June 11, 1979. The instrument used was ZETEX-302A with a 40-foot extension cable.

Readings of 37 selected process tubes were taken at predetermined points. The highest readings were found in the area of the front thermal shield and the front gunbarrel tips. The maximum reading was 251 R/hr with the average being 120 R/hr. Seven localized hot spots were found reading 15 to 78 R/hr; they were located in a random pattern but within 5 feet of the reactor center. The general background in this area inside the reactor is approximately 5 to 10 R/hr.

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TABLE A-8

PROCESS TUBE IN-PLACE DOSE RATE MEASUREMENTS

<u>Reactor</u>	<u>Process Tube No.</u>	<u>Dose Rate at Gunbarrel Tip (R/hr)</u>	<u>Dose Rate 15 ft in from Nozzle (R/hr)</u>
DR Reactor Front Face	0459	28	2.7
	0472	43	3
	0488	35	3.5
	1476	50	Not measured
	2361	70	Not measured
	2374	80	5
	2386	50	Not measured
DR Reactor Rear Face	0659	50	
	0673	70	
	0690	40	
F Reactor Front Face	0459	65	2.5
	0473	75	4
	0488	110	7-24*
	2173	150	4-6 *
F Reactor Rear Face	0659	30	Not measured
	0673	45	Not measured
	0688	27	Not measured
H Reactor Front Face	1873	300	12
	0673	100	8
	0473	65	8

\*In-place dose rate readings went down going away from the gunbarrel tip (1st No.) and then went back up (2nd No.).

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APPENDIX BGRAPHITE SAMPLING OF KW REACTOR

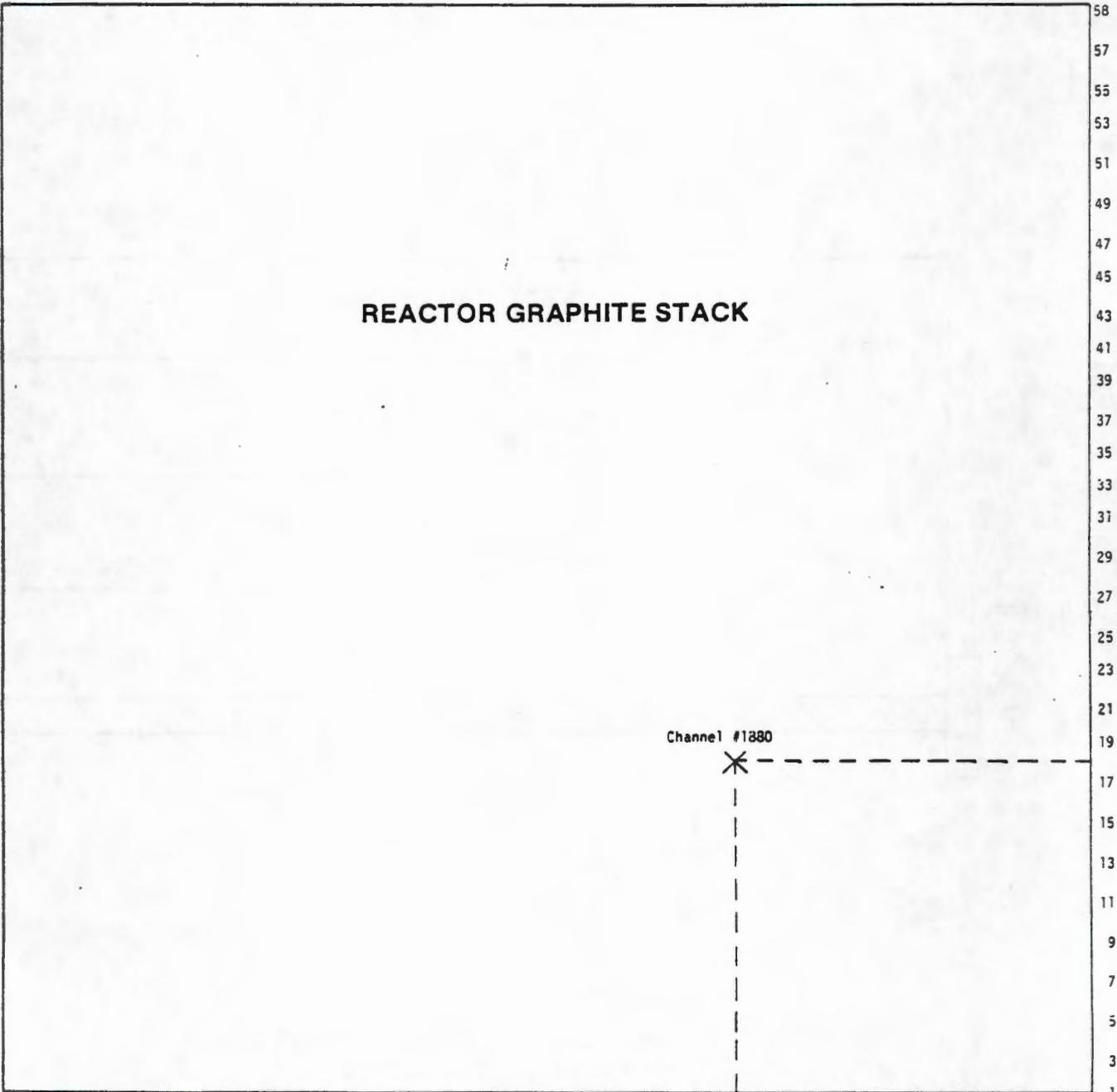
1. Graphite Sample Locations
2. Letter, J. C. Sheppard to J. Stevenson, "C-14 Analysis of Graphite Samples," dated July 24, 1984
3. Letter, W. C. Morgan to J. A. Adams, "Results of Carbon-14 Measurements," dated June 28, 1984
4. Discussion of Sample Analyses
5. Carbon-13 Activation
6. Letter, W. C. Morgan to J. A. Adams, "Results of Nickel-63 Analysis," dated June 7, 1985

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Vertical Rows  
1 thru 58

9 3 1 2 8 7 2 0 4 9 0

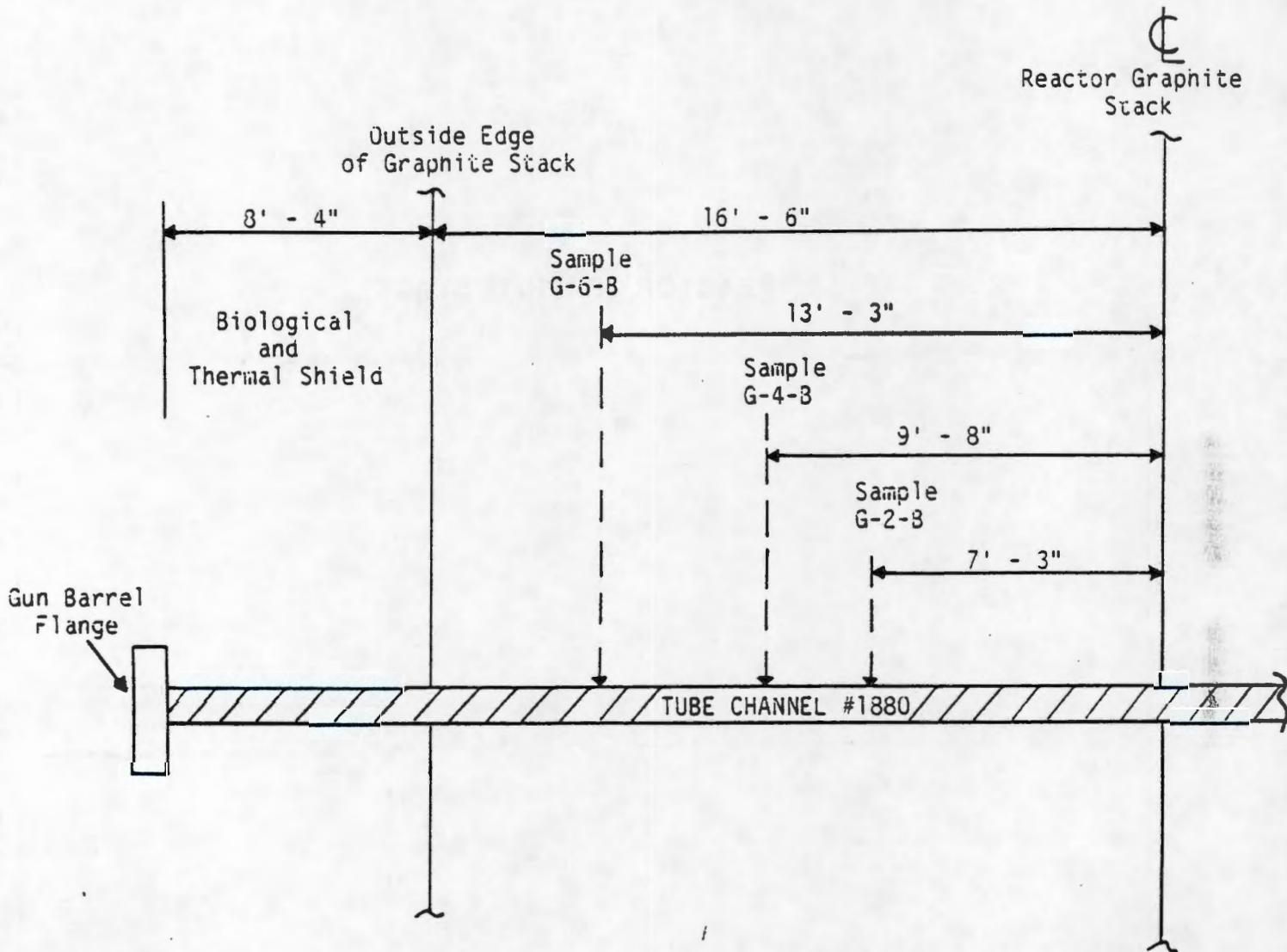


41 43 45 47 49 51 53 55 57 59 61 63 65 67 69 71 73 75 77 79 81 83 85 87 89 91 93 95 97 98

HORIZONTAL TUBE ROWS 41 - 48

**GRAPHITE SAMPLE LOCATION  
KW REACTOR BLOCK  
(FRONT VIEW)**

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**GRAPHITE SAMPLING LOCATIONS  
KW REACTOR BLOCK  
(SIDE VIEW)**

# Washington State University

Department of Chemical Engineering, Pullman, Washington 99164-2710 / 509-335-4332

July 24, 1984

Mr. Jim Stevenson  
UNC Nuclear Industries  
P.O. Box 490  
Rm 430, Federal Building  
Richland, WA 99352

Dear Mr. Stevenson:

Here is our report on the  $^{14}\text{C}$  analysis of your graphite samples.

All samples were burned in oxygen at  $650^{\circ}\text{C}$  to form  $\text{CO}_2$ . Known volumes of  $\text{CO}_2$  were injected in a  $450\text{ cm}^3$  gas proportional counting tube with high purity methane and counted until 1000 to 2000 counts were accumulated. Thus, the counting error ranged between 2.2 and 3.2 percent. Backgrounds of about 2.5 cpm were subtracted off, even though they were obviously negligible.

National Bureau of Standards oxalic acid, the radiocarbon dating standard, was used to obtain the detection efficiency of this detector. At  $25^{\circ}\text{C}$  and 2.00 atmospheres methane, the detector contains 0.442 grams of carbon with a specific activity of 13.6 disintegration per minute per gram of carbon (the equivalent of the NBS oxalic acid). Thus, the disintegration rate should be  $0.442 \times 13.6$  or 6.01 dpm. The observed, long-term average, counting rate for this detector is  $5.10 \pm 0.01$  counts per minute, therefore our detector efficiency is 84.8 percent very close to the value found for similar detectors. In all counts, this efficiency correction was made. With the exception of one sample, all values represent an average of two or more separate counts, sometimes with significantly different sample sizes.

The Table following summarizes our results.

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WSU No.	UNC I.D. No's	cpm/cm <sup>3</sup> CO <sub>2</sub>	dpm/g of carbon
3003	105-DR, DG-33-3	2460	5.91 x 10 <sup>6</sup> (a,b,c)
3004	" , BG-18-3	2227	5.35 x 10 <sup>6</sup>
3005	" , BG-28-3	959	2.30 x 10 <sup>6</sup>
3006	" , DG-28-3	880	2.11 x 10 <sup>6</sup>
3007	" , AG-7-3	360	8.64 x 10 <sup>5</sup>
3016	C-3 Inert	<1	<<2.4 x 10 <sup>3</sup>
3022	105-KW, G-4-B	1327	3.10 x 10 <sup>6</sup>
3023	" , G-6-B	747	1.79 x 10 <sup>6</sup>
3024	" , G-2-B	3396	8.15 x 10 <sup>6</sup>

(a) 
$$\text{dpm/g of carbon} = \text{cpm/cm}^3 \times 22,400 \frac{\text{cm}^3}{\text{mole}} \times \frac{1 \text{ mole}}{12.01 \text{ g carbon}} \times \frac{1}{0.848}$$

(b) Estimated error somewhere between 5 to 10 percent.

(c) Divide by  $2.22 \times 10^6$  to specific activity in microCuries per gram of carbon.

As mentioned above, the estimated errors are in the 5 to 10 percent range. The counting errors, an uncertainty in the counting tube volume and the so-called end-effect contribute to this error. It would take a much more intense research program to obtain higher precision results.

We had expected lower <sup>14</sup>C specific activities, however my calculations suggest that the specific activities reported above may not be too far off. Assuming the following:

$$\sigma_{13} = 0.9 \times 10^{-27} \text{ cm}^2 \text{ } ^{13}\text{C abund} = 0.0111$$

$$\lambda_{14} = 0.693/5740 \text{ yr}^{-1}$$

$$\phi = 2 \text{ to } 3 \times 10^{13} \text{ n/cm}^2 \text{ sec}$$

$$\text{Irrad time} = 10 \text{ to } 30 \text{ years} = \tau$$

$$A_{14} = N_{13} \sigma_{13} \phi \lambda_{14} \tau \text{ dps}$$

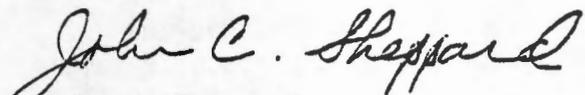
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For 10 years at a flux of  $2 \times 10^{13}$ , we obtain a specific activity of  $7.27 \times 10^5$  dpm/g of carbon.

For 30 years at a flux of  $3 \times 10^{13}$ , the calculated specific activity is  $3.27 \times 10^6$  dpm/gram of carbon. Our calculations are in general agreement with the measurements.

If you have questions, please phone us.

Sincerely,



John C. Sheppard  
Professor of Chemical Engineering  
and Anthropology

JCS/kw

93128720494

Date June 28, 1984

To J. A. Adams

From W. C. Morgan *WCM*

Subject Results of Carbon-14 Measurements

RECEIVED  
JUL 03 1984  
J.A. ADAMS

PO Jackson  
WE Kennedy  
CW Thomas  
LB/File

Our Radiological and Inorganic Chemistry Section obtained the following results for carbon-14 activities from the graphite samples submitted to them.

<u>Reactor</u>	<u>Sample No.</u>	<u><sup>14</sup>C Activity, μCi/g (±2σ)*</u>
--	Control	<3.25 x 10 <sup>-4</sup>
DR	A-G7-2	0.67 ±0.02
DR	B-G18-2	2.36 ±0.04
DR	D-G28-2	2.34 ±0.03
DR	D-G33-2	1.71 ±0.06
KW	G-2A	3.59 ±0.01
KW	G-4A	2.18 ±0.02
KW	G-6A	1.62 ±0.02

Measurement of the <sup>63</sup>Ni activities on samples D-G33-2 and G-A4, should be finished within the next week or two.

\* The error bar limits are counting statistics only, at twice the standard deviation.

WCM/tf

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Discussion of Sample Analyses

The data provided by two independent laboratories, Washington State University (WSU) and Battelle Pacific Northwest Laboratories (PNL), show that the amount of carbon-14 measured varied by sample location and to some extent by the laboratory performing the analysis. For example, if the KW Reactor sample analyses are simply averaged and applied to the entire active region of the graphite stack, values of the carbon-14 are 6400 curies and 5100 curies, respectively for the PNL and WSU analysis values.

Another way to apply the sample analysis data is to assign the values to specific volumes of the reactor. For want of a better way to assign a volume to each sample, the graphite grade (see Appendix C) was used to define the volumes. The total volume of a K Reactor stack is  $1.6 \times 10^9 \text{ cm}^3$  (33.5' x 41' x 41'). The fraction of the stack represented by each grade zone (from Appendix C) are 40%, 30.5%, and 29.5% for the "green", "blue", and "red" zones respectively. Applying the G-2 sample value to the green zone, the G-4 sample value to the blue zone, and the G-6 sample value to the red zone, a carbon-14 value of 6680 curies is found from the PNL analyses. Similarly, a value of 5520 curies of carbon-14 is found from the WSU analysis.

To be conservative, a value of 6700 curies of carbon-14 was assumed for the KW Reactor.

Carbon-13 Activation

Carbon-14 originates from both the nitrogen-14 (n, p) carbon-14 reaction and the carbon-13 (n, gamma) carbon-14 reaction. A simplified calculation of the amount produced by the carbon-13 (n, gamma) carbon-14 reaction was performed. The activity of carbon-14 from the activation of carbon-13 can be represented by the expression:

$$A_{C-14} = f_2 N_0 \lambda / 3.7 \times 10^{10} \text{ curies}$$

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where:

$A_{C-14}$  is the carbon-14 activity per gram of graphite,

$N_0$  is the original number of atoms of carbon-13 per gram of graphite,

$\lambda$  is the mean life of carbon-14,

$f_2$  is the product,  $\phi\sigma t$ , where  $\sigma$  is the capture cross-section of carbon-13,  $\phi$  is the thermal flux, and  $t$  is the irradiation time.

Substituting in the values for the parameters of the equation and assuming a flux of  $10^{14}$  neutrons/cm<sup>2</sup>-s for a time  $t$  of 15 years we have:

$$\lambda = \frac{.693}{\text{half-life of C-14}} = \frac{.693}{5730 \text{ years}} \approx 3.84 \times 10^{-12} \text{ sec}^{-1}$$

$$N_0 = \frac{(.0111)(6.02 \times 10^{23})}{12.01} = 5.56 \times 10^{20} \text{ atoms C-13/gram}$$

$$\sigma = 1.4 \times 10^{-27} \text{ cm}^2$$

$$\phi = 10^{14} / \text{cm}^2\text{-s}$$

$$t = 15 \text{ yrs} \approx 4.73 \times 10^8 \text{ seconds}$$

or

$$A_{C-14} = \frac{f_2 N_0}{3.7 \times 10^{10}} = \frac{(1.4 \times 10^{-27})(10^{14})(4.73 \times 10^8)(5.56 \times 10^{20})(3.84 \times 10^{-12})}{3.7 \times 10^{10}}$$

or

$$A_{C-14} = 3.83 \times 10^{-6} \text{ curies per gram of graphite.}$$

Within the active zone of a K reactor are about  $1.6 \times 10^9$  grams of graphite, so for the total carbon-14 produced by the (n, gamma) reaction of Carbon-13 we have:

Active zone curies =  $(3.83 \times 10^{-6})(1.6 \times 10^9) = 6128$  curies per reactor

A similar computation for the reflector region gives about 120 curies of carbon-14 per reactor. The combined active region plus the reflector region give a total value of about 6250 curies of carbon-14.

93128720498



LB

Date June 7, 1985  
 To J. A. Adams  
 From W. C. Morgan *WCM*  
 Subject Results of Nickel-63 Analysis

Our Radiological and Inorganic Chemistry Section obtained the following results for nickel-63 activities from the graphite core samples. Unfortunately, sample B-G18-2 from DR Reactor was somehow substituted for sample G-A4 from KW Reactor.

<u>Reactor</u>	<u>Sample No.</u>	<u>nCi/g (<math>\pm 2\sigma</math>)*</u>
DR	B-G18-2	28.6 $\pm$ 1.2
DR	D-G33-2	50.9 $\pm$ 1.6

I am also enclosing a copy of a memo from C. W. Thomas, outlining the procedures used to determine the radionuclide activities in the graphite samples.

\* The error bar limits are counting statistics only, at twice the standard deviation.

ias

Enclosure

93128720499

Date August 27, 1984

File/LB

To W. C. Morgan

From C. W. Thomas



Subject

Enclosed in the attached tables are the procedures used in identifying the radionuclide and their concentrations in graphite samples you recently sent us.

Table 1 is the procedure used in determining the radionuclides present and some of their characteristics. Gamma spectrometric measurements showed the samples to consist mainly of  $^{60}\text{Co}$ ,  $^{154}\text{Eu}$ ,  $^{137}\text{Cs}$ , and  $^{133}\text{Ba}$ . Recount after ignition showed the same isotopes with insignificant reduction in concentrations. Beta absorption studies showed essentially a single beta energy of 0.15 MeV. After ignition less than 10% of the beta activity remained.

Table 2 shows the procedure used to determine the radionuclides in the graphite samples that had a beta energy of 0.15 MeV. The results from this procedure confirmed the beta activity was essentially all  $^{14}\text{C}$ .

Table 3 shows the procedure used to separate  $^{63}\text{Ni}$ . The leaching efficiency for removing  $^{63}\text{Ni}$  from the graphite samples was determined by measuring the leaching efficiency of  $^{60}\text{Co}$ . These data showed the leaching efficiency was 81.0% for sample B-18 and 89.9% for D-33. The radiochemical yield for recovery for  $^{63}\text{Ni}$  was determined by tracing with activated  $^{65}\text{Ni}$ . The nickel yield for B-18 was 86.5% while the yield for D-33 was 75.5%. The beta absorption studies confirmed the  $^{63}\text{Ni}$  activity.

If you have any questions concerning the data and/or the procedures used, don't hesitate to call.

CWT:dlc

Encl.

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TABLE 1

## Gamma and Beta Particle Characteristics Procedure.

An aliquot of sample was weighed and placed on a stainless steel dish. The gamma spectro was obtained by measuring on an intrinsic germanium detector. A beta energy analysis was obtained by counting this sample in thin window gas flow beta proportional counter using various thickness aluminum absorbers. The sample was then ignited using a butane burner. The residual material was again gamma and beta counted.

TABLE 2

## Carbon-14 Procedure

Sample aliquot was placed in stainless steel boat with a small amount of copper oxide and inserted into a quartz tube which was placed in a tube furnace. A stream of oxygen was passed over the sample and the gaseous effluent was bubbled through a barium hydroxide trap. The tube furnace was slowly raised to  $\sim 500$  centigrade.

The barium carbonate from the trap was filtered and measured for  $^{14}\text{C}$ .

TABLE 3

## Nickel-63 Procedure

An aliquot of sample was weighed and gamma counted for  $^{60}\text{Co}$ . The sample was then placed in hot 6M HCl, containing carriers and finely powdered by crushing with a glass rod. The sample was leached 3 times. The residue filtered and gamma counted. The leachates were combined and  $^{65}\text{Ni}$  was added. The nickel was separated using dimethylglyoxime with citric acid complexing. The nickel was electroplated on a stainless steel disc and the  $^{65}\text{Ni}$  measured for chemical yield in a gamma spectrometer and the  $^{63}\text{Ni}$  measured in a windowless gas flow proportional beta counted AC shielded with NaI(Tl) detectors. Nickel-63 was verified by beta absorption measurements.

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Pacific Northwest Laboratories

Internal Distribution

Date February 24, 1986

To R. A. Winship

From W. C. Morgan *WCM*

Subject Estimated Chlorine-36 Concentration in the Graphite of the Old Hanford Reactors

JA Adams  
 MD Freshley  
 JA Hall  
 RG Schreckhise  
 ML Smith  
 File/LB

SUMMARY

Samples of 6 grades of nuclear graphites have been analyzed to determine the concentrations of impurities which could be precursors of chlorine-36. For these 6 graphites, residual chlorine is the only significant precursor. Production of  $Cl^{36}$  from the  $(N,\gamma)$  activation of  $Cl^{35}$  has been calculated, assuming that the analyses are representative of the bars used in construction of the old Hanford reactors. If all the original chlorine remains in the graphite, the estimated  $Cl^{36}$  activity in the reactor graphite is as follows:

Reactor:	B	D	F	DR	H	C	KW	KE
$Cl^{36}$ , curies:	42	34	33	54	68	85	271	306

INTRODUCTION

Chlorine-36, which decays by emission of a low-energy beta ray, has a half-life of  $3.1 \times 10^5$  years; thus, it has the potential of becoming a major contributor to the residual radioactivity of nuclear wastes stored for long periods of time. Chlorine-36 can be produced by thermal-neutron  $(n,\gamma)$  activation of  $Cl^{35}$ ; however, it can also be produced by thermal-neutron  $(n,\alpha)$  activation of  $K^{39}$ , or fast-neutron  $(n,p)$  activation of  $Ar^{36}$ . Chlorine is thought to constitute a minor impurity in graphite; but  $Cl^{35}$  can also be produced by the decay of  $S^{35}$ . The relative importance of these separate paths will be quite dependent on the relative concentrations of the impurities; and, the relative concentration can be different for different graphites.

In order to determine the concentrations of elements which could be precursors of  $Cl^{36}$ , samples of six representative graphite grades were selected and analyzed. Table 1 lists the graphites and the pertinent impurity concentrations. Grades KSO and CSO are thermally purified<sup>(a)</sup> graphites, made from different petroleum cokes; KCF and CSF are similar to KSO and CSO, except that they were halogen-purified after the thermal-purification step. Grade GBF is similar to CSF, except that it was not thermally purified; and, SGBF was produced by the same process as GBF, using a different petroleum coke.

(a) "Thermally purified" was used to designate a grade graphitized at a higher temperature than was normal practice.

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February 24, 1986  
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TABLE 1. Data on Graphite Samples

Grade Designation	Heat Number	Density, g/cm <sup>3</sup>	Total Ash, ppm	Impurity Concentration, ppm		
				Cl	S	K
KSO	327	1.717	470	3.9	10.4	0.4
KCF	69	1.652	4	6.6	8.9	nd
CSO	236	1.644	520	2.2	23.	0.2
CSF	602	1.663	7	20.	12.	0.02
GBF	1304	1.692	27	7.0	23.	0.04
SGBF	200	1.605	9	7.8	20.	nd

nd = not detected

Specimens were prepared by sawing a one inch slice from the interior of bars of 4 x 4 in. cross section; both sawed faces were dressed off with a carbide cutter, and care was taken to avoid contamination of the specimens. Densities were determined on the 4 x 4 x 1 inch specimens. Then a representative sample was obtained by drilling nine 0.75 in. diameter holes in each slab using a carbide drill; this produced between 80 and 90 grams of fine powder, which was well mixed before portions of it were taken for analysis.

Total ash was determined by combusting the remainder of the slab (in a covered Pt boat) at 1000°C in air; depending on the weight of the remaining portion of the slab, detection limits ranged between 0.25 and 0.3 ppm. Combustion of the slab allowed the ash pattern to be viewed; from the patterns we can confirm that the powder samples are, indeed, representative of the entire cross section of the bars from which they were cut. The concentrations of Cl and S were determined by Ion Chromatography after combusting the powder samples in an oxygen bomb; detection limits were determined to be 0.3 to 0.5 ppm of Cl, and 0.5 to 0.7 ppm S, using this equipment. The concentration of K was determined by Inductively Coupled Plasma Emission Spectroscopy after combusting the powder samples in pure oxygen and dissolving the ash. The detection limit for K with this equipment is 0.3 µg/ml of solution; with the sample sizes used, this is about 0.07 ppm.

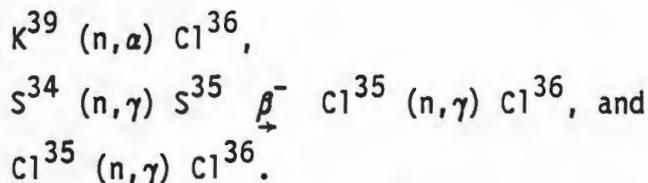
#### Production of Chlorine-36

Four activation reactions were previously identified as potential sources for Cl<sup>36</sup>; these reactions are as follows.

The high-energy neutron (n,p) activation of Ar<sup>36</sup>, and the thermal-neutron activation paths:

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Argon-36 represents only 0.337% of natural argon; and most of the argon in the reactor atmosphere resulted from in-leakage of air, of which argon constitutes 0.93%. Assuming an average concentration of  $N_2$  in the reactor atmosphere of 2%, and a 20% void volume in the graphite, the average concentration of  $Ar^{36}$  in voids in the graphite would be only about  $10^{25}$  ppm at operating temperature. Thus,  $Ar^{36}$  activation can be ignored under the present situation.

The concentration of K in these samples is also much too low to contribute significantly to the production of  $Cl^{36}$ .

The loss of  $S^{34}$  is described by the differential equation:

$$- \frac{d[S^{34}]}{dt} = [S^{34}] \sigma \phi \quad (1)$$

Where:  $[S^{34}]$  is the concentration of  $S^{34}$  at time  $t$ ,

$\sigma$  is the  $(n, \gamma)$  cross section (0.23b)\* and

$\phi$  is the thermal neutron flux intensity.

The solution to equation (1) can be written:

$$[S^{34}] = [S^{34}]_0 e^{-\sigma \phi t} \quad (2)$$

Where:  $[S^{34}]_0$  is the concentration of  $S^{34}$  originally in the graphite, and the other quantities are defined above.

Sulfur-34 constitutes only 4.21% of natural S; thus, the highest concentration of  $S^{34}$  in any of the graphites is ( $23 \text{ ppm} \times 0.0421 =$ ), 0.99 ppm. Table 2 shows that the maximum value of  $\int_0^t \phi dt$  (at the center of KE Reactor) for any of the graphites is  $5.51 \times 10^{22} / \text{cm}^2$ ; thus, from equation (2) one can calculate that the  $S^{34}$  activation and decay scheme could increase the  $Cl^{35}$  content by not more than

$$0.99 (1 - e^{-0.01267}) = 0.012 \text{ ppm.}$$

Thus, we can conclude that  $S^{34}$  activation can safely be ignored under the present circumstances.

\* One barn is  $10^{-24} \text{ cm}^2$ .

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TABLE 2. Calculation of Thermal Neutron Fluence

Reactor	Final Oper. Power, MW	Total Power Gen., 10 <sup>6</sup> MWd	Equivalent FPD <sup>(1)</sup>	Fluence ( $\phi t$ ), 10 <sup>22</sup> /cm <sup>2</sup> <sup>(2)</sup>
B	1960	9.98	5092	2.20
D	2005	9.89	4933	2.13
F	1935	8.09	4181	1.81
DR	1925	6.95	3610	1.56
H	1955	7.80	3990	1.72
C	2310	11.80	5108	2.21
KW	4400	22.14	5043	5.23
KE	4400	23.37	5311	5.51

(1) Full power days of operation.

(2) Peak thermal flux at full power was taken as  $1.2 \times 10^{14}/\text{cm}^2 \text{ sec}$  for KE and KW,  $5 \times 10^{13}/\text{cm}^2 \text{ sec}$  for all other reactors.

The  $^{35}\text{Cl}$  ( $n, \gamma$ ) activation reaction is, thus, the only significant source of  $^{35}\text{Cl}$  for the graphites in the old Hanford reactors. The destruction of  $^{35}\text{Cl}$  by this activation reaction can be described by the following differential equation:

$$-\frac{d[^{35}\text{Cl}]}{dt} = [^{35}\text{Cl}] \sigma_c \phi \quad (3)$$

Where:  $[^{35}\text{Cl}]$  is the concentration of  $^{35}\text{Cl}$  at time  $t$ ,  $\sigma_c$  is the cross section (43b) for the  $^{35}\text{Cl}$  ( $n, \gamma$ )  $^{35}\text{Cl}$  activation reaction, and  $\phi$  is the thermal neutron flux.

The solution to equation (3) is:

$$[^{35}\text{Cl}] = N_0 e^{-\sigma_c \phi t}$$

Where:  $N_0$  is the concentration of  $^{35}\text{Cl}$  originally in the graphite.

The half-life of  $^{36}\text{Cl}$  ( $3.1 \times 10^5$  years) is sufficiently long that we can ignore the loss of  $^{36}\text{Cl}$  during a period of 40 years; thus, the production of  $^{36}\text{Cl}$  can be equated to the loss of  $^{35}\text{Cl}$ , to obtain the following equation:

$$[^{36}\text{Cl}] = N_0 (1 - e^{-\sigma_c \phi t}) \quad (4)$$

Where:  $[^{36}\text{Cl}]$  is the concentration of  $^{36}\text{Cl}$  at time  $t$ .

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### REACTOR ZONES AND GRAPHITE GRADES

#### B, D, and F Reactors

The graphites used in the three oldest Hanford reactors were produced at Morganton, NC, and Clarksburg, WV, during 1943 and 1944. Samples of the available petroleum cokes and coal-tar pitches had been analyzed for ash, boron, vanadium, titanium, iron, and calcium; on the basis of these tests, 2 cokes (Kendall and Cleves) and Barrett #30 pitch were chosen as the standard raw materials. A second pitch (Barrett Chicago 7H0) was also used, because it contained only 0.4 ppm boron as opposed to 1.0 ppm boron in "Standard" (Barrett #30) pitch; however, Chicago 7H0 was available only in limited quantities. Table 3 lists the three grades of graphite and average ash, boron, and vanadium content, determined on 50 tons of each graphite.

TABLE 3. Data on Graphites Produced for B, D, and F Reactors

<u>Grade</u> <sup>(1)</sup>	<u>Coke</u>	<u>Pitch</u>	<u>Ash, %</u>	<u>Boron, ppm</u>	<u>Vanadium, ppm</u>
KC	Kendall <sup>(2)</sup>	Chicago	0.05	0.45	15
KS	Kendall	Standard	0.05	0.50	15
Cs	Cleves <sup>(3)</sup>	Standard	0.06	0.60	25

(1) Some WS (Whiting-Standard) was also produced; but, it was not used in the fueled regions of these reactors.

(2) Produced from a Pennsylvania paraffin-base crude oil by the Kendall Refining Company, Bedford, PA.

(3) Produced from an asphalt-base crude oil at the Cleves, OH, refinery of Gulf Oil Company.

The original plan was to construct the stacks using four grades of graphite; the purest grade was to be installed in the central region, and successively less pure graphites used at increasing distance from the center. Accordingly, the stacks were divided into four zones, which were designated by color: Green, White, Blue, and Red; the Green zone being at the center of the stack, and the Red zone completely outside the fueled region. Problems encountered during production resulted in a change of plans, and the final distribution of grades is shown in Table 4.

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Zone	Zone Volume, % of Stack	Graphite Grades		
		B Reactor	D Reactor	F Reactor
Green	12	KC	KC	71% KC, 29% CS
White	25	78% KS, 13% KC, 9% CS	CS	64% KS, 36% CS
Blue	46	CS	CS	CS
Red	17	Misc. (1)	Misc. (2)	Misc. (2)

(1) Believed to be primarily grade WS graphite.

(2) Believed to be primarily low quality KC and KS; however, grade CS and other graphites may have been used.

Exact dimensions of the zones are not known; however, the shape of the zones followed the general shape of the pile, and the zone boundaries were chosen to approximately conform to planes of equal flux intensity, assuming an unflattened flux profile. The fixed lengths of the graphite bars precludes a strict definition of a zone boundary; however, Table 5 shows zone dimensions which were chosen on the basis of the above, and other, considerations.

TABLE 5. Assumed Zone Boundaries in B, D, and F Reactors

Zone	Outside Dimension, ft			Zone Volume	
	Width	Height	Length	ft <sup>3</sup>	% of Stack
Green	18	18	14	4536	12.5
White	26	26	20	8984	24.8
Blue	34	34	26	16536	45.6
Red	36	36	28	6232	17.2

#### DR, H, and C Reactors

The production of KC, KS, and CS graphites for these three reactors commenced at Morganton, NC, in August of 1947 and extended until March 1949; during this period, several significant changes were made in the purification process.

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at high-temperature with carbon tetrachloride ( $\text{CCl}_4$ ) and dichlorodifluoromethane ( $\text{CCl}_2\text{F}_2$ , "Freon-12"). Following a series of tests on graphites purified at Bay City, purification furnaces were set up at Morganton, and production of the halogen-purified grades KCF, KSF, CSF, etc., began in January 1948. The "F" designation was given to thermally-purified grades which were halogen-purified, "Finished," in a separate operation carried out at lower temperatures than those attained during graphitization. Initially,  $\text{CCl}_4$  vapors (using  $\text{N}_2$  as the carrier gas) was used both during the heat-up cycle to  $2000^\circ\text{C}$ , and during the cool-down cycle, below  $2000^\circ\text{C}$ ; F-12 was used above  $2000^\circ\text{C}$ , primarily to remove boron (and, as it turned out, the rare earths). However, it was soon learned that excessive amounts of Cl were retained in the graphite unless it was swept out of the furnace at temperatures not less than  $1000^\circ\text{C}$ ; nitrogen was used as the sweep-gas for all subsequent "F"-processing.

In order to produce a graphite which would not expand as rapidly as CSF does when irradiated at low temperatures, and to reduce production costs, a series of experimental runs were made in which the thermal-purification, graphitization, step was omitted. The nuclear purity of the experimental bars proved to be equal to that of normal CSF bars; therefore, the facilities at Morganton were converted to allow simultaneous purification and graphitization to take place at about  $2450^\circ\text{C}$ . Graphites produced by this process commenced in November of 1948; only Cleves Coke and Standard pitch were used, and the new grade was designated GBF (Gas-Baked, F-purified) graphite. The use of  $\text{N}_2$  as the carrier and sweep-gas continued until February 11, 1949, only a few weeks before production ceased.

As a result of these changes in the manufacturing process, the graphite grades used in the different zones varies significantly for these three reactors. In addition, size of the zones was changed for H and C; however, the zones in DR are identical to those in B, D, and F reactors. Table 6 shows the approximate zone dimensions in H and C reactors.

TABLE 6. Assumed Zone Boundaries in H and C Reactors

Zone	Outside Dimensions, ft			Zone Volume	
	Width	Height	Length	ft <sup>3</sup>	% of Stack
Green	16	16	14	3584	9.9
White	24	24	20	7936	21.9
Blue	30	30	26	11880	32.7
Red	36	36	28	12888	35.5

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Table 7 shows the approximate composition of the zones in DR, H, and C reactors; there is some uncertainty in the actual percentages for the various grades, because only the number of "heats" were recorded. A "heat" is one furnace load; the number of bars in a heat varied for the different processes, and varied with time for a given process. Thus, an extensive review of the production records would be required to obtain a better estimate of the actual percentages by grade designation.

TABLE 7. Approximate Graphite Allocation in DR, H, and C Reactors

Zone	Graphite Grades <sup>(1)</sup>		
	DR Reactor	HR Reactor	C Reactor
Green	86% KCF, 13% CSF, 1% KSF	68% CSF, 17% GBF, 15% KCF	GBF
White	40% KC, 20% KS, 14% CS, 15% CSF, 11% KCF	87% CSF, 12% GBF, 1% KCF	GBF
Blue	66% CS, 22% KC, 12% KS	87% CS, 13% GBF	GBF
Red	71% CS, 17% KC, 12% KS	48% CS, 45% KS, 7% Misc. <sup>(2)</sup>	93% CS, 7% KS

(1) Grade designations which indicate position in a heat (such as KCNF, CSNF, CSQ, etc.) are tabulated as part of the primary grade (KCF, CSF, CS, etc.).

(2) Listed as "Group II," no other identification; these may be early F-processed bars with high-chlorine content.

#### KW and KE Reactors

Most of the graphite used in KW and KE reactors was manufactured by National Carbon Company<sup>(a)</sup> during 1953 and 1954; the coke that was used was produced from a mid-continent crude oil at the Lockport, IL, refinery of the Texas

(a) Now: Carbon Products Div. of Union Carbide Corp.

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Co. (a) Graphite destined for the moderator was GBF processed and designated TSGBF. Speer Carbon Co. (b) produced a small amount (used in the filler layers of the Green zone in KE) from the same coke, and manufactured to the same specifications as TSGBF; the Speer grade was designated SGBF. Graphite for the reflector region (Red zone) was thermally-purified and designated TS-AGOT.

Table 8 lists the graphites allocated to the different zones in KW and KE.

TABLE 8. Graphite Allocation for KW and KE Reactors

Zone	% of Stack	Graphite Grades	
		KW	KE
Green	40	68% TSGBF, 32% CSGBF <sup>(1)</sup>	57% TSGBF, 43% SGBF
Blue	30.5	63% TSGBF, 37% CSGBF <sup>(2)</sup>	TSGBF
Red	29.5	55% TS-AGOT, 37% CS, 5% KC <sup>(3)</sup>	85% TS-AGOT, 15% TSGBF <sup>(4)</sup>

(1) Includes 10% CHF; CHF is a CS graphite, extruded at Clarksburg in 1944 and GBF processed in 1948.

(2) Includes about 1% CHF and 0.5% CSF.

(3) Remaining 3% is CSGBF plus a few bars of TSGBF and KCF.

(4) Includes a few bars of CSGBF.

#### CALCULATIONS

Equation 4 can be rewritten in the form:

$$[Cl^{36}]_p = N_0 (1 - e^{-\sigma_c f \phi t}) \quad (5)$$

Where:  $[Cl^{36}]_p$  is  $Cl^{36}$  concentration (ppm) at time  $t$ , at position  $p$  in the reactor (ppm)

$N_0$  is  $Cl^{35}$  concentration (ppm) at time  $t = 0$ ,

$\sigma_c$  is the activation cross section ( $43 \times 10^{-24} \text{ cm}^2$ ),

(a) Now: Texaco, Inc.

(b) Now: AIRCO Carbon.

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$f$  is the ratio of the fluence sustained at position  $p$ , to the maximum fluence,  $\phi_t$ , sustained in the reactor ( $\phi_t$  is listed in Table 2 for each reactor).

$N_0$  can be calculated from the measured concentrations of chlorine listed in Table 1, the natural abundance of  $Cl^{35}$  (75.77%) in chlorine, and the relative amounts of each grade of graphite in a reactor zone (Tables 4, 7, and 8). The fraction  $f$  can be calculated from the approximate zone dimensions (Tables 5 and 6, <sup>(a)</sup> assuming a flattened side-to-side, and cosine front-to-rear, flux distribution. Tables 9 and 10 list the calculated values for  $N_0$  and  $f$  for each reactor zone.

TABLE 9. Original Concentrations of Chlorine-35 by Reactor Zone

Reactor	Chlorine-35 Concentration, ppm			
	White	Green	Blue	Red
B	2.96	2.84	1.67	2.96
D	2.96	1.67	1.67	2.96
F	2.58	2.49	1.67	2.96
DR	8.34	6.37	2.10	2.04
H	6.80 (1)	7.28 (1)	2.14	3.31 (2)
C	(3)	5.30	5.30	1.76 (4)
KW	(3)	(3)	5.70	2.55 (4)
KE	(3)	(3)	5.97	3.40 (4)

- (1) The dih of the CSF bars was too high to justify use of 20 ppm chlorine, therefore, the CSF in these zones was assumed to be 10 ppm.
- (2) "Group-II" was assumed to have high chlorine content; assumed 20 ppm Cl as an average.
- (3) White and Green zones were combined for C Reactor; White, Green, and Blue were combined for KW and KE.
- (4) Chlorine content of TS-AGOT assumed the same as in KSO, 3.9 ppm.

- (a) KW<sub>3</sub> and KE are treated as having only 2 zones, a Green/Blue zone of 39675 ft<sup>3</sup>, encompassing (essentially) the central 54 x 54 tube rows, and a Red zone of 16629 ft<sup>3</sup>.

R. A. Winship  
February 24, 1986  
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TABLE 10. Fractional Fluence (f) by Reactor Zone

Reactor	Ratio of Fluence/Peak Fluence in the Reactor			
	White	Green	Blue	Red
B, D, F, and DR	0.826	0.668	0.429	0.035
H	0.826	0.676	0.566	0.147
C	---	0.723	0.566	0.147
KW and KE	---	---	0.637	0.301

Table 11 lists the  $\text{Cl}^{36}$  concentration in each reactor zone, calculated using equation 5.

TABLE 11. Chlorine-36 Concentration by Reactor Zone

Reactor	Chlorine-36 Concentration, ppm			
	White	Green	Blue	Red
B	1.61	1.33	0.56	0.10
D	1.57	0.76	0.54	0.09
F	1.22	1.01	0.47	0.08
DR	2.67	1.76	0.53	0.05
H	3.11	2.86	0.73	0.34
C	--	2.63	2.20	0.23
KW	--	--	4.34	1.25
KE	--	--	4.65	1.73

The total activity of  $\text{Cl}^{36}$  in each reactor zone can be calculated as follows:

$$A_i = [\text{Cl}^{36}]_i W_i N \lambda / M R \quad (6)$$

Where:  $A_i$  is the activity (curies) of  $\text{Cl}^{36}$  in zone  $i$ ,

$[\text{Cl}^{36}]_i$  is the average concentration of chlorine-36 ( $10^{-6}$  x ppm) in the graphite of zone  $i$ ,

$W_i$  is the weight (grams) of graphite in zone  $i$ ,

$N$  is Arogadro's number ( $6.023 \times 10^{23}$ /mole),

$\lambda$  is the decay constant for  $\text{Cl}^{36}$  ( $7.09 \times 10^{-14}$ /sec),

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February 24, 1986  
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M is the molecular weight for  $\text{Cl}^{36}$  (35.45 g/mole), and

R is the conversion factor from disintegrations per second to Curies ( $3.7 \times 10^{10}$ /sec).

$W_i$  can be further defined as follows:

$$W_i = V_i (1-v_i) C \rho \quad (7)$$

Where:  $V_i$  is the volume ( $\text{ft}^3$ ) of zone  $i$ ,

$v_i$  is the fractional void volume in zone  $i$ ,

C is the constant (0.02832) which converts  $\text{ft}^3$  to  $\text{m}^3$ ,

$\rho$  is density of the graphite ( $\text{Mg}/\text{m}^3$ ).

The central  $657 \text{ m}^3$  region of DR Reactor includes a void volume of  $39 \text{ m}^3$ ; this is a void fraction,  $v_i$ , of about 6%. This has been applied as a density correction to the zones (or parts of zones) in B, D, F, DR, H, and C Reactors; a void fraction of 8% was assumed for the corresponding regions in KW and KE. The average densities of the graphites used in each zone have been documented for all reactors, except B, D, and F. The density of KC and KS used in these reactors was assumed to be the same as that used in the White and Green zones of B Reactor; the density of grade CS was assumed to be the same as that of the CS used in DR Reactor. Table 12 lists the densities by zone for each of the reactors.

TABLE 12. Average Graphite Densities  
by Reactor Zone

Reactor	Average Graphite Density, $\text{Mg}/\text{m}^3$			
	White	Green	Blue	Red
B	1.656	1.693	1.667	1.700
D	1.656	1.667	1.667	1.700
F	1.659	1.685	1.667	1.700
DR	1.718	1.687	1.666	1.672
H	1.661	1.652	1.660	1.693
C	---	1.648	1.659	1.656
KW	---	---	1.644	1.663
KE	---	---	1.677	1.675

Combining equations 6 and 7, we can write the following equation for total  $\text{Cl}^{36}$  activity per zone:

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February 24, 1986  
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$$A_i = [Cl^{36}]_i V_i \rho_i (1-v_i) C N \lambda / M R \quad (8)$$

Table 13 lists the calculated  $Cl^{36}$  activity of the reactors by zone.

TABLE 13. Activity of Chlorine-36 by Reactor Zone

Reactor	Chlorine-36 Activity, Curies				
	White	Green	Blue	Red	Total
B	10.5	17.5	13.5	1.0	42.4
D	10.2	9.9	12.9	0.9	33.9
F	8.0	13.3	11.2	0.8	33.3
DR	18.0	23.1	12.7	0.5	54.3
H	16.0	32.5	12.5	6.7	67.7
C	--	43.3	37.4	4.4	85.1
KW	--	--	240.1	31.2	271.3
KE	--	--	262.4	43.5	305.9

#### DISCUSSION

It should be noted that the purity of graphite varies from bar-to-bar, as well as from heat-to-heat. (a) Moreover, the impurities are not uniformly distributed within a bar; they tend to cluster in small areas. Thus, there is no guarantee that a slice from a single bar is representative of that grade of graphite. Table 14 illustrates the type of differences which can be expected.

TABLE 14. Comparison of Selected Impurity Concentrations

Reactor	Zone	Graphite Grade	Impurity Concentration, ppm				
			Boron	Vanadium	Titanium	Iron	Calcium
B	Green	KC	0.4	16	26	800	100
D	Green	KC	0.6	16	27	800	100
-	---	KSO	4.0	11	8	6	212
B	Blue	CS	0.6	45	23	600	50
D	White	CS	0.5	45	23	600	50
D	Blue	CS	0.6	45	23	600	50
F	Blue	CS	0.6	16	25	800	100
--	---	CSO	3.2	12	8	3	135

(a) A "heat" is a graphitization, or purification, furnace load.

R. A. Winship  
February 24, 1986  
Page 14

The large difference in iron content between the CS and KC, produced during 1943-1944, and the samples of CSO and KSO, produced during 1947-1949, is surprising. However, the ash residue from both samples was a light-beige color, not the rust-red color of ash from samples high in iron. Another unanticipated result was that 33 and 31 ppm of sulfur was retained in the ash from the CSO and KSO samples, respectively; these are 1.5X and 3X the amount of sulfur found in the smaller samples combusted in the oxygen bomb.

The high (20 ppm) concentration of chlorine in the CSF sample is believed not to be representative of normal CSF graphite; during the early production of CSF, treatment with  $\text{CCl}_4$  was continued until the furnace had cooled to fairly low temperatures. Chlorine concentrations as high as 35 ppm were measured on bars from some of these early production runs; however, the retention of Cl considerably reduced the nuclear purity as measured by dih. <sup>(a)</sup> Because the CSF used in H Reactor has nearly the same dih as the GBF used in C Reactor, there cannot be large differences in Cl concentration; therefore, a Cl concentration of 10 ppm was assumed for the CSF used in H Reactor. Conversely, the low dih for the CSF used in DR Reactor (compared to that of the CSF in H Reactor or the KCF in DR Reactor) justifies the use of the measured 20 ppm Cl for CSF in DR Reactor. (The difference, about 0.18 dih, could be produced by 12 ppm Cl.)

One other fact should be explicitly mentioned: about one-half the residual chlorine was removed when a bar of CSF was baked at  $1400^\circ\text{C}$  for 2 hours; however, baking at  $200^\circ\text{C}$  had no effect on dih. It is possible that the chlorine content in the reactors may have been reduced during operation.

#### REFERENCE

Nichols, P. F., and E. M. Woodruff. 1962. "Nuclear Properties," Nuclear Graphite, R. E. Nightingale, editor, Academic Press, NY, pp. 67-85.

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(a) Delta-in-hours. See: Nichols and Woodruff (1962).

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APPENDIX D

SOURCE OF TRITIUM IN GRAPHITE

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JA Adams  
WL Templeton  
File/LB

Date October 3, 1985  
To M. L. Smith  
From W. C. Morgan   
Subject Extrapolation of DR Reactor Core  
Activities to Other Reactors

Although the graphites used in the oldest reactors were produced by the same manufacturing process, impurity contents of these graphites can vary significantly both within a grade of graphite and between grades. Very little data is available on elemental composition of the impurities; however, some appreciation of the differences can be gained from the changes in nuclear purity. Nuclear purity, measured in delta-in-hours (dih), is most heavily influenced by the concentrations of boron, the rare earths, vanadium, and titanium; however, for the same starting materials and purification process, total impurity content seems to have been well correlated with dih.

In the older piles, graphite was sorted by dih; that with the highest dih (highest purity) was used in the central "Green Zone," slightly less pure material was used in the "White Zone," even less pure in the "Blue Zone," and the remainder was relegated to the outer "Red Zone." For B Reactor, graphite in the Green Zone averaged +0.255 dih; in D, 75% averaged +0.309 dih (the remainder averaged +0.203 dih); and in F, 29% averaged +0.334 dih, with the remainder measuring +0.309 dih (however, a "new batch" of KS became available with a dih of +0.622, and some of this was used in the White Zone).

In contrast, graphite in the Green Zone at DR averaged +0.946 dih, and graphite in the Red Zone averaged only slightly lower dih (-0.30 vs. -0.25) than some of that used in the White Zone at B Reactor. More important, all of the graphite used in the Green Zone, and some of that in the White Zone at DR was F-Processed; the F purification process reduced impurity levels by an order of magnitude, or more, even on the early batches.

Variability in source of the cokes, hence, variability in impurity concentrations, introduces another potential problem in extrapolating measurements at DR to the other reactors. Part of this uncertainty could be removed by elemental analysis of the graphites used to construct the different reactors. I have archived one or more bars of most of the graphite grades used in the Hanford reactors; sections of these bars could be analyzed to determine relative concentrations of the important elements.

I should also mention that the tritium in the graphite probably is primarily a result of the reaction of fast neutrons with nitrogen:  $N^{14} + n \rightarrow C^{12} + H^3$ , which is the source of  $H^3$  in our atmosphere. Thus, like  $C^{14}$ , its distribution in the graphite may be dependent on impurity concentrations, temperature gradients which existed during plant operation, etc.; however, because of the difference in the energies of the neutrons required for the reactions, the ratio of  $C^{14}$  to  $H^3$  should vary with proximity to the fuel.

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APPENDIX E

RATIO OF <sup>59</sup>Ni TO <sup>63</sup>Ni

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Ratio of  $^{59}\text{Ni}$  to  $^{63}\text{Ni}$

The ratio of  $^{59}\text{Ni}$  to  $^{63}\text{Ni}$  may be represented by the expression:

$$\frac{^{59}\text{Ni}}{^{63}\text{Ni}} = \frac{\lambda_{59}}{\lambda_{63}} \cdot \frac{\sigma_{58}}{\sigma_{62}} \cdot \frac{N_{58}}{N_{62}} \quad \text{Equation A}$$

where:

$$\lambda_{59} \text{ is the mean life of } ^{59}\text{Ni} = \frac{.693}{7.6 \times 10^4} \text{ yr}^{-1}$$

$$\lambda_{63} \text{ is the mean life of } ^{63}\text{Ni} = \frac{.693}{100} \text{ yr}^{-1}$$

$$\frac{N_{58}}{N_{62}} \text{ is the ratio of target isotopes of Nickel} = \frac{68.27}{3.59}$$

$$\sigma_{58} \text{ is thermal absorption cross-section of } ^{58}\text{Ni} = 4.6 \text{ b}$$

$$\sigma_{62} \text{ is thermal absorption cross-section of } ^{62}\text{Ni} = 14.5 \text{ b}$$

Substituting into Equation A

$$\frac{^{59}\text{Ni}}{^{63}\text{Ni}} = \frac{9.12 \times 10^{-6}}{6.93 \times 10^{-3}} \cdot \frac{4.6}{14.5} \cdot \frac{68.27}{3.59} = 0.0079$$

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APPENDIX F

CONTAMINATED FACILITY OUTSIDE THE REACTOR BLOCK  
AND FUEL STORAGE BASINS

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ESTIMATED RADIONUCLIDE INVENTORY OF GROUND  
DISPOSAL FACILITIES UNDER A MOUND OVER  
A REACTOR ASSUMING A 3 TO 1 SLOPE

The in situ option using a mound with a radius of 210 feet and a 3 to 1 slope is assumed. The ground disposal facilities under the mound are as follows: 116-B-3 Pluto Crib; 116-B-4 dummy decontamination tank drain; 116-KE-1 115 Crib; 116-KW 115 Crib and the 105-C Ball 3X disposal silos.

The curie burdens estimated for these facilities are listed below:

<u>Facility Identification</u>	<u>Radionuclides</u>	<u>Estimated Curies* (March 1985)</u>
116-B-3 Pluto Crib	<sup>3</sup> H	0.22
	<sup>90</sup> Sr	0.02
	<sup>137</sup> Cs	0.14
	<sup>152</sup> Eu	0.26
	<sup>154</sup> Eu	0.03
	<sup>238</sup> U	0.0003
	<sup>238</sup> Pu	0.0001
	<sup>239</sup> Pu	0.0006
116-B-4 Dummy Decon Drain	<sup>60</sup> Co	0.02
116-KE-1 Crib	<sup>3</sup> H	80
	<sup>14</sup> C	110
	Others	1
116-KW-1 Crib	<sup>3</sup> H	80
	<sup>14</sup> C	110
	Others	1
105-C Ball 3X Silos**	<sup>60</sup> Co	80
	<sup>63</sup> Ni	2

\*Inventories based on data in UNI-946 and decayed to March 1, 1985.

\*\*This is the only ground disposal facility that may be disturbed should the one piece reactor dismantlement alternative be selected.

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APPENDIX G

HAZARDOUS MATERIAL INVENTORY

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**UNC NUCLEAR INDUSTRIES**

A UNC RESOURCES Company

P.O. Box 490  
Richland, Washington 99352

Telephone 509/376-7411

## Memorandum

To: J. A. Adams

Date: December 20, 1985

From: D. H. Doerge *DH Doerge*

Subject: HAZARDOUS MATERIALS INVENTORY

An inspection of the deactivated reactor buildings was conducted to identify and inventory any hazardous materials which remained in the facilities following deactivation. Materials found consisted mainly of lead brick and lead sheet which was used for shielding and lesser amounts of lead wool, cadmium sheets, mercury switches, oils and possibly solvents.

The inspection consisted of a walk-through of each facility with personnel who were very familiar with areas which were suspect and relatively inaccessible. Lighting conditions were very poor and most of the inspection was made using portable lighting (flashlights). Due to the poor lighting and inaccessibility of some areas some materials may have been missed but it is felt that any overlooked material would have a very small impact on the total inventory.

For consistency all materials are reported in pounds unless noted otherwise. Lead bricks were assumed to weigh approximately twenty-five pounds each and other forms were estimated by volume (cubic feet). Volumes were converted to weight at seven hundred pounds per cubic foot.

Ball caves and other caves located on top of the reactors were assumed to contain lead equal to one-third (1/3) of the total volume of the cave. Where possible, lead brick used for shielding was counted and portions of shield walls which were not visible were estimated.

Oil reservoirs for elevator equipment are located on the top level in each facility. Contents of the reservoirs were not investigated. Equipment is assumed to contain oil in the gear cases.

Some areas in 105-KE and 105-KW were not accessible at this time and should be inspected during the site preparation phase of decommissioning. These areas are located on the back side of the reactors, ball rooms, rear face, etc.

DHD/jrf

cc: JA Hall  
MC Hughes  
BW Mathis  
RF Potter  
RA Winship  
LB

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## HAZARDOUS MATERIALS INVENTORY

FACILITY 105-BPAGE 1 OF 1

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE		COMMENTS
				YES	NO	
9/25/85	Front far side 0'	Lead	700 lbs	x		Lead brick
9/25/85	Front far side -9'	Lead-Cadmium	500 lbs		x	Monitoring tube
9/25/85	Front far side -9'	Lead	7,000 lbs	x		Cave (lead brick)
9/25/85	Front far side -9'	Lead	1,000 lbs	x		Lead brick
9/25/85	Front far side X-1	Lead	500 lbs		x	Shield plugs
9/25/85	Top of Reactor	Lead	3,500 lbs	x		7 rod tip shields
9/25/85	Rod Room - outer	Lead	625 lbs	x		Lead brick
9/25/85	Rod Room - inner	Lead	1,000 lbs	x		Lead brick
9/25/85	Sample rooms	Lead	2,000 lbs	x		Lead shot in sample trays
9/25/85	Rear face tool room	Lead	50 lbs	x		Lead sheet
9/25/85	Rear face +10'	Lead	5 lbs	x		Lead wool
9/25/85	Elevator Eqpt Level	Lead	1 ea	x		Mercury switch

UNI-3714







9 3 1 2 8 7 2 0 5 2 8

## HAZARDOUS MATERIALS INVENTORY

FACILITY 105-FPAGE 1 OF 1

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE		COMMENTS
				YES	NO	
10/8/85	Far side -9'	Lead	60 lbs	x		Lead plate
10/8/85	Far side X-1	Lead	1,400 lbs	x		Lead brick shielded box
10/8/85	Far side X-1	Lead	60 lbs	x		Lead sheet on pipe
10/8/85	Far side X-1	Lead	60 lbs	x		Lead plate
10/8/85	Far side X-2	Lead	1,000 lbs	x		Lead brick - scattered
10/8/85	Far side X-2	Lead	120 lbs	x		Lead sheet on pipe & lead plate
10/8/85	Far side X-2	Lead	20 lbs	x		Lead wool
10/8/85	Far side X-2	Lead	500 lbs	x		Lead brick around tube
10/8/85	Far side X-2	Cadmium	25 lbs	x		Neutron shield bonnets (2)
10/8/85	Far side X-2	Cadmium	5 lbs	x		Cadmium sheet
10/8/85	Top of Reactor	Lead	4,000 lbs	x		Rod tip shields (6)
10/8/85	Top of Reactor	Lead	4,000 lbs	x		Lead brick (airway to rear face)
10/8/85	Top of Reactor	Lead	9,000 lbs		x	Lead brick (2 caves)
10/8/85	Sample rooms	Lead	5,000 lbs	x		Lead shot in sample trays
10/8/85	Rod room - inner	Lead	125 lbs	x		Lead brick
10/8/85	Rod room - outer	Lead	2,100 lbs	x		Shields plugs
10/8/85	Near side -9'	Lead	300 lbs	x		Lead brick shielded box
10/8/85	Front face -0'	Lead	25 lbs	x		Lead brick on duct
10/8/85	Near side -9'	Cadmium ?	2 ea	x		Splines

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## HAZARDOUS MATERIALS INVENTORY

FACILITY 105-IIPAGE 1 OF 1

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE		COMMENTS
				YES	NO	
10/8/85	Far side -9'	Lead	250 lbs	x		Lead brick & sheet
10/8/85	Far side X-2	Lead	200 lbs	x		Lead brick & sheet
10/8/85	Far side X-3	Lead	500 lbs	x		Lead brick
10/8/85	Far side X-3	Cadmium	20 lbs	x		Sheet around experiment
10/8/85	Top of Reactor	Lead	4,200 lbs	x		Rod tip shields
10/8/85	Top of Reactor	Lead	2,500 lbs	x		Lead brick
10/8/85	Top of Reactor	Lead	4,000 lbs		x	Cave
10/8/85	Top of Reactor	Lead	4,000 lbs		x	Ball cave
10/8/85	Far side	Lead	40,000 lbs		x	Shield door (equip. hatch)
10/8/85	Sample rooms	Lead	4,000 lbs	x		Lead shot in sample trays
10/8/85	Near side -12'	Lead	750 lbs	x		Lead brick
10/8/85	Near side -12'	Lead	1,000 lbs	x		Lead sheet around piping
10/8/85	Rod room - inner	Lead	800 lbs	x		Lead brick & lead sheet
10/8/85	Rod room - outer	Lead	2,100 lbs	x		Shield plugs & shield
10/8/85	Near side stairway	Mercury	--	x		2 Mercury switches
10/8/85	Near side -12'	Mercury	--	x		3 Mercury switches
10/8/85	Near side -12'	Solvent	--	x		Tank (contents unknown)

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HAZARDOUS MATERIALS INVENTORY

FACILITY 105-KW

PAGE 1 OF 1

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE		COMMENTS
				YES	NO	
12/19/85	-9 Landing	Lead	25 lbs.	X		Lead Brick
"	Outer Rod Room	Lead	500 lbs.	X		Lead Brick
"	Outer Rod Room	Lead	100 lbs.	X		Lead Sheet
"	Inner Rod Room	Lead	2500 lbs.	X		Lead Brick
"	Inner Rod Room	Lead	10,000 lbs.		X	Rod Tip Shield
"	X-1 Level	Lead	750 lbs.	X		Lead Brick
"	X-1 Level	Lead		X		6 Shielded Pigs
"	X-2 Level	Lead	300 lbs.	X		Lead Sleeve
"	X-2 Level	Lead	5000 lbs.	X		Lead Brick
"	Top of Reactor	Lead	250 lbs.	X		Lead Brick
"	Top of Reactor	Mercury	47 ea.	X		Mercury Switches
NOTE: The far side rooms were not inspected. Area is boarded up to discourage entry.						

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# UNC NUCLEAR INDUSTRIES



A UNC RESOURCES Company

PO Box 490  
Richland, Washington 99352

UNI-3714  
RECEIVED  
MAR 10 1986  
Telephone 509/876-7411  
MILLER

## Memorandum

To: J. A. Adams/D. H. Doerge Date: March 5, 1986

From: L. E. Denton *L. E. Denton*

Subject: HAZARDOUS MATERIALS INVENTORY

Reference: Memorandum, D. H. Doerge to J. A. Adams, same subject, dated March 5, 1986.

Due to the current attitude on mercury and the method of disposal being in place, we have removed the switches listed in the letter. Also, many other switches were located and removed.

They will be disposed of by V. D. Apple. The mercury switches are at 1717-K/100-K Area. Any time we locate or are notified of mercury in a deactivated facility, we will remove it for disposal.

If you have any questions, please contact R. E. Jennings on 3-1440.

LED/bej

cc: RE Jennings  
BW Mathis  
LED-File/LB

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