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Potential Carryover of Alpha Contamination to a Reactor
by Specimens Handled in a Hot-Cell Environment
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ABSTRACT

A study was conducted to determine how much uranium and plutonium contamination is potentially carried over to EBR-II by specimens handled in the HFEF/S Hot Cells. Plutonium levels on contaminated non-irradiated fuel elements were determined by a novel alpha counter, while uranium contamination levels were estimated from the known plutonium levels and the approximate ratio of U/Pu contamination in-cell. Contaminated elements were then subjected to tests, studying the contamination adherence in hot flowing argon gas and in hot molten sodium. These tests were designed to simulate conditions in the transfer devices and in the reactor primary tank.

The experiment results indicate that less than 10^{-4} $\mu\text{g}/\text{cm}^2$ plutonium and less than 0.1 $\mu\text{g}/\text{cm}^2$ uranium are typically carried out of the hot-cell environment by fuel elements, routinely handled and examined in-cell. Probably, none of this contamination is removed by hot argon gas, circulating through the transfer devices, while less than 40% probably remains on elements immersed in the primary tank sodium.

Based on these results, it is estimated that less than 1.7 μg Pu-239 and less than 0.9 mg U-235 remain on reconstituted experiment

surfaces in the EBR-II core. This would imply that plutonium contamination, fixed to reconstituted experiments in the reactor core, is not the source of the background fission product gas activity currently observed in the reactor cover gas. However, fixed U-235 contamination may be the source of this activity.

POTENTIAL CARRYOVER OF ALPHA CONTAMINATION
TO A REACTOR BY SPECIMENS HANDLED IN
A HOT-CELL ENVIRONMENT

A Thesis

Submitted to the Graduate Faculty of the
Louisiana State University and
Agricultural and Mechanical College
in partial fulfillment of the
requirements for the degree of

Master of Science

in

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by

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DEDICATED TO

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ABSTRACT

A study was conducted to determine how much uranium and plutonium contamination is potentially carried over to EBR-II by specimens handled in the HFEF/S Hot Cells. Plutonium levels on contaminated non-irradiated fuel elements were determined by a novel alpha counter, while uranium contamination levels were estimated from the known plutonium levels and the approximate ratio of U/Pu contamination in-cell. Contaminated elements were then subjected to tests, studying the contamination adherence in hot flowing argon gas and in hot molten sodium. These tests were designed to simulate conditions in the transfer devices and in the reactor primary tank.

The experiment results indicate that less than 10^{-4} $\mu\text{g}/\text{cm}^2$ plutonium and less than 0.1 $\mu\text{g}/\text{cm}^2$ uranium are typically carried out of the hot-cell environment by fuel elements, routinely handled and examined in-cell. Probably, none of this contamination is removed by hot argon gas, circulating through the transfer devices, while less than 40% probably remains on elements immersed in the primary tank sodium.

Based on these results, it is estimated that less than 1.7 μg Pu-239 and less than 0.9 mg U-235 remain on reconstituted experiment surfaces in the EBR-II core. This would imply that plutonium contamination, fixed to reconstituted experiments in the reactor core, is not the source of the background fission product gas activity currently observed in the reactor cover gas. However, fixed U-235 contamination may be the source of this activity.

CHAPTER I

INTRODUCTION

Experimental Breeder Reactor II (EBR-II)* is an important facility for irradiations testing under the Atomic Energy Commission's Liquid Metal Fast Breeder Reactor (LMFBR) Program. The major goal of this program is to develop a prototype LMFBR for commercial use.⁽¹⁾ The irradiations tests are essential for the selection of materials and the development of reliable future nuclear fuels.

The Hot Fuel Examinations Facilities (North and South*) play a major role in EBR-II's irradiations testing program. Here, irradiated specimens are remotely examined behind the shielding of thick concrete walls. Many experiments are also rebuilt in HFEF after interim examinations before being transferred to EBR-II for further irradiation. HFEF/S has been recycling fuel and experiments to EBR-II for over ten years, while HFEF/N will begin operations in late 1974. HFEF/S has been primarily a nondestructive testing facility, but a few fueled experiments have been destructively examined there. Many experiments will be destructively examined in HFEF/N, so there will be a possibility of having much more fissionable material carried over to EBR-II on the surfaces of reconstituted experiments.

There are three fundamental ways in which more tramp (or undesirable) uranium and plutonium surface contamination could affect

*See Appendix A for background information concerning EBR-II and HFEF.

reactor operation: (1) Contamination could be deposited in the devices which transport experiments to the reactor tank, (2) it could wash off into the primary sodium and could remain suspended or settle out on primary tank components, or (3) it could remain fixed to experiment surfaces in the core.

Experiments are transferred from HFEEF to EBR-II by way of the Interbuilding Transfer Coffin (IBC) and the Fuel Unloading Machine (FUM). In both of these devices, hot argon gas is circulated to heat the experiment, so contamination could be removed by hot flowing argon gas while an experiment is enroute to EBR-II. Eventually, levels could build up to a point where contact maintenance on the FUM or IBC could be hazardous. Contamination settling out on components in the primary tank could also present a contact maintenance problem. And finally, tramp fissionable material suspended in the core sodium or fixed to surfaces in the core could cause monitoring problems for EBR-II.

Potentially, the most serious effect that tramp uranium and plutonium contamination can have on EBR-II is associated with the reactor monitoring systems. The reactor cover gas is continually monitored for fission product activity. The presence of a large amount of fission product gas in the reactor cover gas is indicative of a leaking fuel element or capsule. Fissionable material in the reactor core will also produce fission product gas, so substantial amounts of tramp uranium and plutonium in the core sodium or on experiment surfaces could reduce the effectiveness of these monitors.

The germanium-lithium argon scanning system (GLASS) is one of three on-line detectors monitoring the reactor cover gas for fission product gas. ⁽³⁾ This monitor currently indicates that the equivalent of 2 - 3 mg tramp U-235 is in the core. It is estimated that approximately 15 mg equivalent U-235 in the core would compromise the monitoring effectiveness of GLASS. ⁽⁴⁾

According to recent analyses, there are less than 2 ppb uranium and less than 0.004 ppb plutonium in the primary sodium. ⁽⁵⁾ This implies that there are less than 10 μ g fissionable material distributed homogeneously in the core sodium. Smears of components removed from the primary tank have failed to detect either uranium or plutonium as surface contaminants. Therefore, fissionable material suspended in the core sodium is probably not the source of the observed cover gas activity.

There are two possible explanations for the observed fission product activity: (1) fission product gases are diffusing through stainless steel cladding and subsequently diffusing to the reactor cover gas, or (2) there is fissionable material fixed to surfaces in the reactor core. ⁽²⁾ The only surfaces in the core are fuel element and sub-assembly hardware surfaces. Hence, uranium and plutonium fixed to experiment surfaces could be a source of this fission product gas activity.

The amount of tramp uranium and plutonium contamination which presently commutes between HFEF/S and EBR-II is unknown. The amount of contamination removed from experiment surfaces by hot flowing argon gas in the transfer devices and by molten sodium in the primary tank is also

unknown. The objectives of this study are: (1) to determine how much tramp uranium and plutonium contamination is typically carried out of HFEF/S by experiments, (2) to estimate how much of this contamination is removed by hot flowing argon gas in fuel handling equipment such as FUM and IBC, (3) to estimate how much of this contamination is removed by molten sodium in the primary tank, and (4) to estimate how much contamination is fixed to experiment surfaces in the core.

CHAPTER II

REVIEW OF PAST RESEARCH

Attempts have been made to determine typical levels of uncontained plutonium present in both the air cell and the argon cell of HFEF/S. The prime tool used for surveying plutonium levels on component surfaces in cell has been smearing. Numerous sets of smears have been made in the cell environment in the last few years, the most comprehensive set being recorded in June 1973 when Cook and Holson⁽⁶⁾ conducted an experiment to determine the amount of plutonium suspended in the argon cell atmosphere before and during destructive examination of a plutonium-bearing safety test loop.

However, these studies did not help to quantify typical levels of plutonium and uranium that leave the hot cell environment on experiment surfaces. For example, smearing is only a qualitative indicator of typical levels of removable contamination.^(7,8,9) It is not an acceptable method for obtaining quantitative information concerning the carryout problem. Although past smear studies do show that plutonium is present in both the air and argon cell, they do not indicate how much is typically being carried over to EBR-II. There is apparently nothing known about how much uranium is typically carried out of cell. It is quite possible that U-235 is as bad a contaminant as is Pu-239. Hence, this part of the problem cannot be ignored.

There has been very little research in the area of how well uranium and plutonium contamination adheres to stainless steel surfaces.

Although there is some active research currently in progress at the Dounreay Experimental Reactor in Scotland on the adherence of uranium to stainless steel surfaces immersed in molten sodium, results have not been published in the open literature. Ebersole and Brunson performed an experiment in which a known amount of uranium was plated onto a stainless steel surface. ⁽¹³⁾ The sample was then immersed in a hot sodium bath, after which it was removed and the remaining uranium was measured. In this experiment, all of the deposited uranium was removed by the sodium bath.

In summary, past studies offer little quantitative information on carryout or adherence of tramp uranium and plutonium contamination on specimens examined in a hot-cell environment.

used to measure microgram quantities of uranium because of its low specific activity. However, a standard should be established for

CHAPTER III

EXPERIMENT PROCEDURES AND RESULTS

The experimentation was divided into two major areas: (1) experiments measuring levels of plutonium and uranium contamination carried out by stainless steel specimens, and (2) experiments studying the adherence of this contamination to these specimens. Conditions in the primary tank and transfer devices were used to help determine experiment procedures.

Evaluation of Detection Methods

Various chemical methods have been widely used in the past to measure microgram quantities of uranium and plutonium. (10,11,12) These methods are accurate and adaptable to highly radioactive specimens. However, there are several disadvantages for using chemical methods to measure contamination levels. For instance, chemical techniques are costly and time consuming. This makes analysis of a large number of samples impractical, and statistically meaningful data would be difficult to accumulate. Furthermore, the contamination would have to be removed from the specimens for analysis. This would greatly complicate studies on contamination adherence in various fluids as there would be no way to compare contamination levels before and after a test.

Detection of the alpha particles emitted by plutonium offers the possibility of quantifying contamination levels in situ. The counting method is inexpensive, requires little time to make a measurement, and is very sensitive, at least for plutonium. Alpha counting could not be

used to measure microgram quantities of uranium because of its low specific activity. However, if a U/Pu ratio could be established for cell contamination, this ratio could be used to make realistic estimates of uranium contamination levels from the plutonium count rate.

An alpha counter which could potentially detect plutonium on fuel element surfaces was designed and tested (see Appendix C). The fundamental element in the detection system was a cylindrical gas flow ionization chamber, which utilized argon as the counting gas. This detector was novel in that the anode was a typical fast reactor fuel element. Pulses, produced by radiation ionizing the argon gas, were amplified, discriminated against and counted. In a series of out of cell tests, the detection efficiency of this counter was determined to be about 49% over a range of alpha activities from 2000 to 50,000 dpm, and was unaffected by beta-gamma backgrounds up to 100 mR/hr.

A test was conducted to determine the feasibility of operating the detector remotely. Remote operation would be necessary in order to monitor irradiated elements for plutonium. The detector chamber was inserted into the hot-cell environment, while the gas flow system and counter electronics were placed out-of-cell. Gas and electronics lines were fed through the cell wall to the detector chamber. The chamber had to be less than 10 feet from the counter electronics as signal cables which were longer than 10 feet produced background pulses which exceeded the counter lower discriminator level. The test showed that the electronics and gas lines could be remotely attached with master slave

manipulators. In addition, fuel elements could be readily loaded into and unloaded from the detector chamber.

The second part of this in-cell test determined if the counter could be used to monitor plutonium on irradiated fuel elements. In the original test procedure, an irradiated fuel element was to have been brought up next to the outside wall of the detector and its effects on the detector noted. The final test was to have been an alpha count of an irradiated fuel element. These tests, however, were never conducted in the air cell. With parameters fixed at values obtained in the out-of-cell testing, the detector failed to operate.

As argon began to flow into the detector, the background pulse heights rose from a level of 1 volt to a level of 10 volts. The background gradually returned to its initial level about two hours after argon flow had been terminated. Various remedies were tried to correct the problems. A new polyethylene cap was made to insure good clip contact. A polyethylene tube was inserted into the gas feed line to remove all possible grounding problems created by the stainless steel gas line. Various detector gas mixtures were tried. Pure argon gas was replaced by a 90% argon, 10% methane mixture, a mixture that is often used in gas flow counters because of its favorable quenching characteristics. A 75% helium, 25% argon mixture was also tried. Finally, amplifier gain, discriminator settings and high voltages were varied in an effort to find some stable operating point. However, all of these efforts were unsuccessful.

It was, therefore, inferred that the in-cell detector could not tolerate the radiation levels of the air cell. A test was performed out-of-cell to determine what radiation levels the alpha detector could tolerate. A detector chamber, similar to the in-cell chamber, was placed in the radiation field of a 100 curie Ir-192 source. The detector and source were placed in a lead-lined room, normally used for gamma and x-radiography. The source was exposed by cranking it out of a lead shield to the end of a radiography snake. The resulting field was essentially of 4-pi geometry. A Jordon Radector Detector, model ACB-550-S, measured the approximate dose rate to the detector. The Radector probe was positioned on the alpha detector base plate, while both the probe signal cable and the alpha detector preamplifier cable were fed to their respective counting circuits outside the room. The dose rate to the detector was varied by moving the snake head away from the alpha detector. The response of the alpha detector to these various field strengths was then observed on an oscilloscope.

The experiment showed that the 2-inch alpha detector could not tolerate a gamma dose rate of greater than about 10 R/hr. As Table I illustrates, for dose rates above this level, background pulse heights exceeded the lower discriminator setting of 2.0 volts. In general, background pulse heights were roughly proportional to the cube root of the dose rate.

As a consequence, this detector could not be used to monitor contamination levels on irradiated fuel elements. The following course of experimentation was, therefore, chosen to study the problem of how much

contamination is carried out-of-cell by fuel elements. Non-irradiated contaminated elements were subjected to tests which simulated conditions in the transfer devices and in the primary tank. The alpha counter, mentioned above, was used to monitor plutonium levels on element surfaces before and after each of these tests. The corresponding uranium levels were then estimated from the approximate ratio of U/Pu contamination in-cell, which was obtained from chemical analysis.

A Preliminary Study Using Steel Blocks

TABLE I

The objective of this study was to obtain an order of magnitude estimate of the detector background as a function of dose rate.

Detector Distance from Source (inches)**	Observed Detector Dose Rate (R/hr.)	Detector Background Pulse Height (volts)
80	18	2
64	28	3
32	90	4
14	400	6

*Lower discriminator voltage levels = 2.0 volts.

**Distance measured from center of Radector probe to snake head.

A Preliminary Study Using Stainless Steel Blocks

The objectives of this experiment were: (1) to obtain an order of magnitude estimate of uranium and plutonium contamination picked up in-cell by stainless steel surfaces from fallout, (2) to obtain an order of magnitude estimate of uranium and plutonium contamination picked up in-cell by stainless steel surfaces from handling, (3) to obtain qualitative information concerning the buildup of surface contamination with time in-cell, and (4) to obtain an approximate ratio of U/Pu contamination levels in-cell.

The test specimens in this experiment were twenty stainless steel cylindrical blocks (see Figure 1). Each block was 1-inch high by 3/4-inch diameter and made of type 304 stainless steel. A removable spring-steel bail was inserted into the top of each block for handling purposes. In this way, a block could be handled in-cell without touching the experimental surface. Each block was polished and cleaned with acetone before being introduced into the cell.

The twenty blocks were divided into five sets of four. A number and a letter were etched on the bottom of each block for identification. The number indicated a particular block set; the letter indicated a particular block within a set (e.g. 1A was block A in set 1).

A stainless steel box was used to insert the blocks into the cell. The box was subdivided into twenty separate compartments by an ice tray type divider. The compartments were labeled similar to the blocks, with rows being numbered 1 to 5 and compartments within a row being lettered A to D. Thus, each block had a specified location in the box. The box

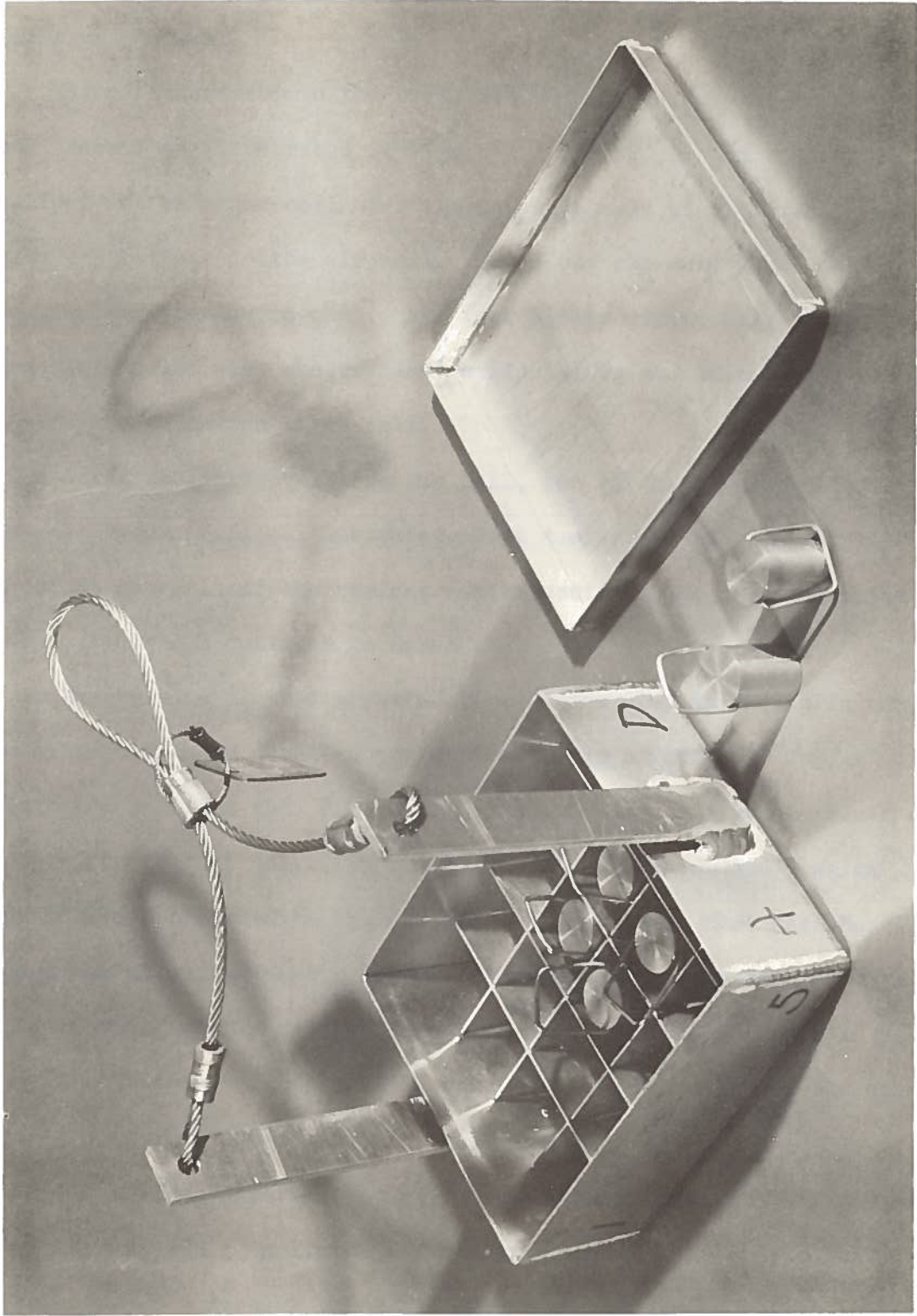


FIGURE 1. The Stainless Steel Blocks and Transfer Box Used in the Study.

had a removable lid to prevent contamination of the blocks in the transfer process.

After transfer into the cell, the block sets were moved to the cell locations illustrated in Figure 2. Sets 2, 3, and 4 measured fallout in the argon cell over varying intervals of time, while set 1 measured fallout in the air cell. Set 5 was the only block set intentionally handled in-cell. All four blocks within this set were grossly handled with master slave manipulators and dusted with a cleanup brush in the argon cell.

After their removal from the cell, the blocks were transferred to the Argonne National Laboratory-West Analytical Section for chemical analysis of the surface contamination. The procedures for the analyses are outlined in Appendix B.

The results of this experiment are summarized in Tables II and III. Referring to these data, contamination levels of about $10^{-5} \mu\text{g}/\text{cm}^2$ plutonium and less than $0.04 \mu\text{g}/\text{cm}^2$ uranium were observed on block surfaces from fallout only. About $10^{-3} \mu\text{g}/\text{cm}^2$ plutonium and $1 \mu\text{g}/\text{cm}^2$ uranium were observed on the surfaces of the blocks that were handled. There was no significant buildup of contamination with time in-cell. This is most readily seen by comparing data from fallout sets 1, 2, and 3. (The fallout data from set 4 were lost in the course of the experiment due to the mishandling of these blocks.)

The maximum observed ratio of U/Pu contamination adhering to block surfaces was 878. Explicit contamination ratios were only obtained for blocks in set 5 because the small amount of uranium on the fallout

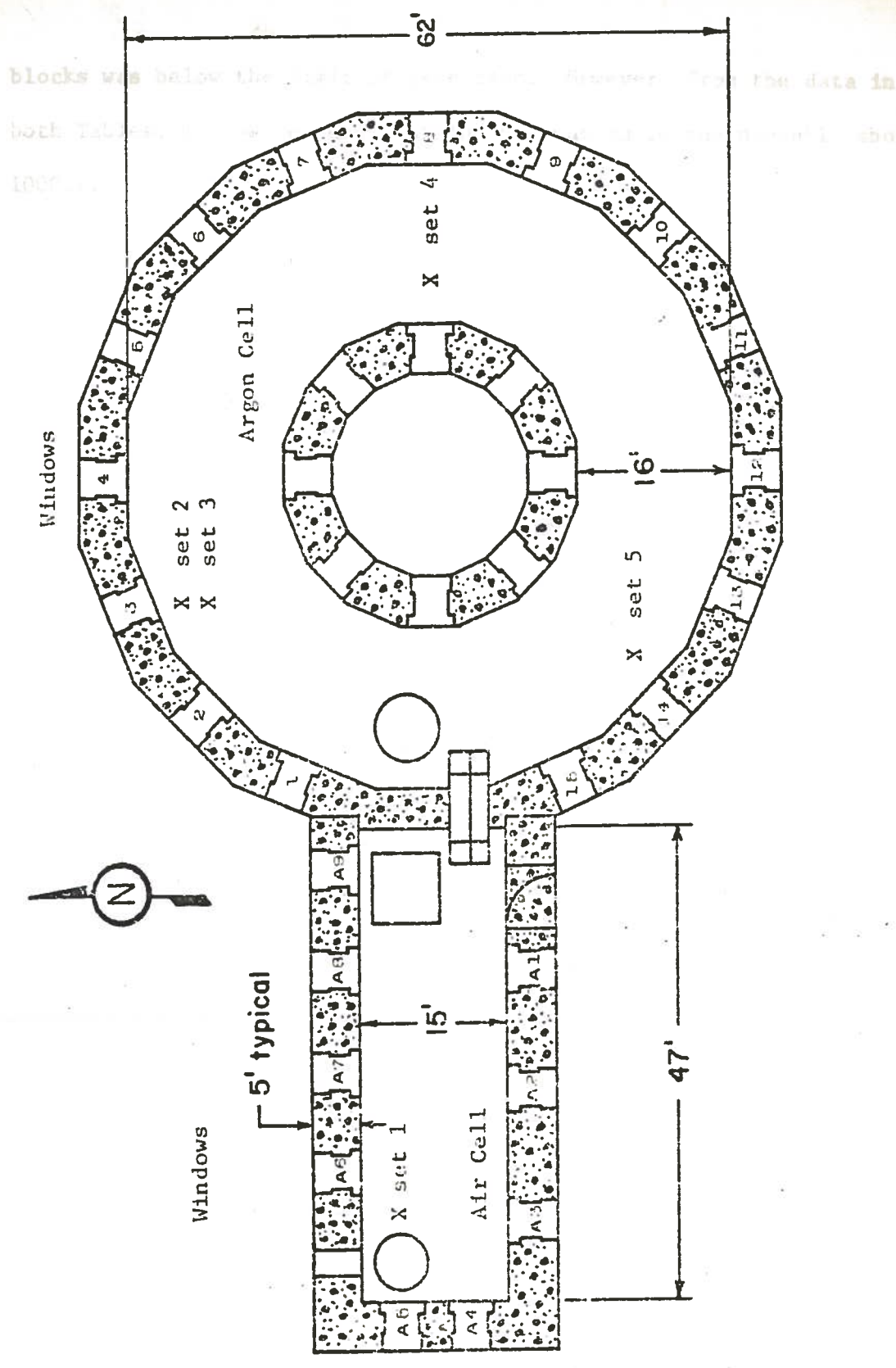


FIGURE 2. Location of Block Sets in the Air and Argon Cells.

blocks was below the limit of detection. However, from the data in both Tables, it can be seen that the maximum ratio was probably about 1000:1.

Sample No.	Average #	Standard Deviation	Observed (n/csm ²)
1	0.04	0.04	0.04
2	0.04	0.04	0.04
3	0.04	0.04	0.04
4	0.04	0.04	0.04
5	0.04	0.04	0.04
6	0.04	0.04	0.04
7	0.04	0.04	0.04
8	0.04	0.04	0.04
9	0.04	0.04	0.04
10	0.04	0.04	0.04
11	0.04	0.04	0.04
12	0.04	0.04	0.04
13	0.04	0.04	0.04
14	0.04	0.04	0.04
15	0.04	0.04	0.04
16	0.04	0.04	0.04
17	0.04	0.04	0.04
18	0.04	0.04	0.04
19	0.04	0.04	0.04
20	0.04	0.04	0.04
21	0.04	0.04	0.04
22	0.04	0.04	0.04
23	0.04	0.04	0.04
24	0.04	0.04	0.04
25	0.04	0.04	0.04
26	0.04	0.04	0.04
27	0.04	0.04	0.04
28	0.04	0.04	0.04
29	0.04	0.04	0.04
30	0.04	0.04	0.04
31	0.04	0.04	0.04
32	0.04	0.04	0.04
33	0.04	0.04	0.04
34	0.04	0.04	0.04
35	0.04	0.04	0.04
36	0.04	0.04	0.04
37	0.04	0.04	0.04
38	0.04	0.04	0.04
39	0.04	0.04	0.04
40	0.04	0.04	0.04
41	0.04	0.04	0.04
42	0.04	0.04	0.04
43	0.04	0.04	0.04
44	0.04	0.04	0.04
45	0.04	0.04	0.04
46	0.04	0.04	0.04
47	0.04	0.04	0.04
48	0.04	0.04	0.04
49	0.04	0.04	0.04
50	0.04	0.04	0.04
51	0.04	0.04	0.04
52	0.04	0.04	0.04
53	0.04	0.04	0.04
54	0.04	0.04	0.04
55	0.04	0.04	0.04
56	0.04	0.04	0.04
57	0.04	0.04	0.04
58	0.04	0.04	0.04
59	0.04	0.04	0.04
60	0.04	0.04	0.04
61	0.04	0.04	0.04
62	0.04	0.04	0.04
63	0.04	0.04	0.04
64	0.04	0.04	0.04
65	0.04	0.04	0.04
66	0.04	0.04	0.04
67	0.04	0.04	0.04
68	0.04	0.04	0.04
69	0.04	0.04	0.04
70	0.04	0.04	0.04
71	0.04	0.04	0.04
72	0.04	0.04	0.04
73	0.04	0.04	0.04
74	0.04	0.04	0.04
75	0.04	0.04	0.04
76	0.04	0.04	0.04
77	0.04	0.04	0.04
78	0.04	0.04	0.04
79	0.04	0.04	0.04
80	0.04	0.04	0.04
81	0.04	0.04	0.04
82	0.04	0.04	0.04
83	0.04	0.04	0.04
84	0.04	0.04	0.04
85	0.04	0.04	0.04
86	0.04	0.04	0.04
87	0.04	0.04	0.04
88	0.04	0.04	0.04
89	0.04	0.04	0.04
90	0.04	0.04	0.04
91	0.04	0.04	0.04
92	0.04	0.04	0.04
93	0.04	0.04	0.04
94	0.04	0.04	0.04
95	0.04	0.04	0.04
96	0.04	0.04	0.04
97	0.04	0.04	0.04
98	0.04	0.04	0.04
99	0.04	0.04	0.04
100	0.04	0.04	0.04

TABLE II

SUMMARY OF BLOCK EXPERIMENT DATA

Block Set No.	In-Cell Location	Time in Cell (days)	Range of Pu Levels Observed		Average Pu Levels Observed ($\times 10^{-5}$ $\mu\text{g}/\text{cm}^2$)	Range of U Levels Observed		Average U Levels Observed ($\mu\text{g}/\text{cm}^2$)
			($\times 10^{-5}$ $\mu\text{g}/\text{cm}^2$)	($\mu\text{g}/\text{cm}^2$)		($\mu\text{g}/\text{cm}^2$)	($\mu\text{g}/\text{cm}^2$)	
1	Air Cell, Window A6	26	2.18 - 8.02		5.37	<0.04		<0.04
2	Argon Cell, Window 3	12	2.00 - 5.66		3.28	<0.04		<0.04
3	Argon Cell, Window 3	26	0.40 - 4.38		3.14	<0.025*		<0.025
4	Argon Cell, Window 9	-	-		-	-		-
5	Argon Cell, Window 14	12	142 - 1130		472	0.72 - 2.6		1.36

*This valve was smaller since a larger sample of the extracted uranium solution was analyzed.

Levels of Contamination Carried out-of-Cell by Typical Fuel Elements

The objectives of this investigation were to determine how much contamination is carried out of the cell by typical fuel elements routinely...

TABLE III
RATIOS OF U/Pu CONTAMINATION ON BLOCKS IN SET 5

Block	Pu/Block (μg)	U/Block (μg)	U/Pu
5A	5.00 x 10 ⁻²	15	300
5B	2.35 x 10 ⁻¹	55	234
5C	7.82 x 10 ⁻²	18	230
5D	2.96 x 10 ⁻²	26	878

Levels of Contamination Carried Out-of-Cell by Typical Fuel Elements

The objectives of this experiment were: (1) to determine how much contamination is typically carried out-of-cell by fuel elements routinely handled and examined in the hot cell environment and (2) to determine if there is an increase in the amount of contamination carried out-of-cell due to storage in the argon cell.

Test elements in the experiment were 75 Mark 1A fuel element jackets (see Figure 3). Each jacket was welded closed at the top, duplicating the geometry of a typical fuel element. Also at the top of each element, a red stripe was painted to prevent confusing a dummy element with an irradiated Mark 1A element. A number from 1 to 75 was stamped for identification on each bottom spade. Each element was cleaned with acetone prior to insertion into the cell.

The experiment consisted of a duplication of the routine handling and examination process that fuel elements undergo in the air and argon cells. The important steps in the experiment were:

- 1) Insertion of the dummy elements into the air cell.
- 2) Transfer of the elements to a Mark 1A fuel element magazine.
- 3) Elements subjected to routine air cell examinations. Examinations performed included: profilometry, visual examinations, and gamma scan (see Appendix A).
- 4) Elements 1 to 25 removed from the air cell.
- 5) The remaining 50 elements transferred to the argon cell for storage.
- 6) Elements 26 to 50 removed from cell after 2 months storage.

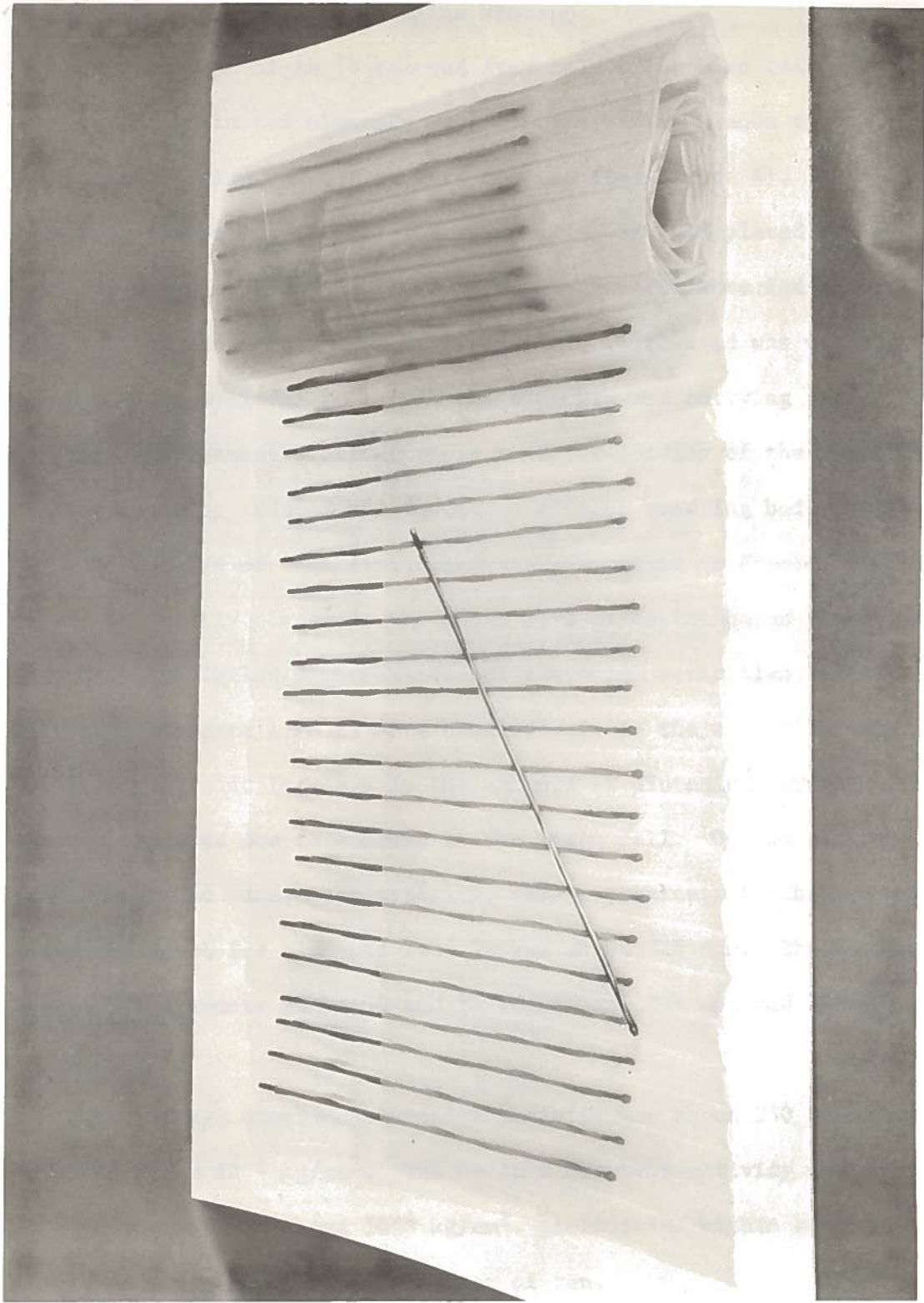


FIGURE 3. The Elements Which Were Routinely Handled In-Cell.

7) Elements 51 to 62 run through the sodium bond tester in the argon cell after 4 months storage.

8) Elements 51 to 75 removed from cell after bond testing.

The contaminated elements were transferred to a hood in the Analytical Section where they were counted (see Figure 4). The alpha detector, elements, and their carrying grid were all placed in the same hood. Gas and electronics lines from the detector were fed to equipment outside the hood. A sheet of 1/2-inch thick lead was wrapped into an open-ended cylinder to shield the elements and carrying grid. This counting arrangement provided ample room for loading of the detector chamber and also reduced the amount of element handling before a count.

The results of this experiment are summarized in Figures 5, 6, and 7. As is readily observed, the three sets of contaminated elements show surprisingly similar distributions of plutonium activities, despite the fact that elements 1 to 25 were handled only in the air cell. There was no significant increase in the amounts of plutonium carried out-of-cell by elements due to storage in the argon cell. On the contrary, elements stored were, statistically, less contaminated. The average plutonium level for elements 1 to 25 was about 326 dpm. The average levels for elements 26 to 50 and 51 to 75 were 204 dpm and 225 dpm, respectively.

The average observed plutonium activity was about 250 dpm/element, or about $2.5 \times 10^{-5} \mu\text{g}/\text{cm}^2$. The maximum observed activity was about 860 dpm/element, or about $10^{-4} \mu\text{g}/\text{cm}^2$. Activities within each set of 25 elements varied by about a factor of ten.

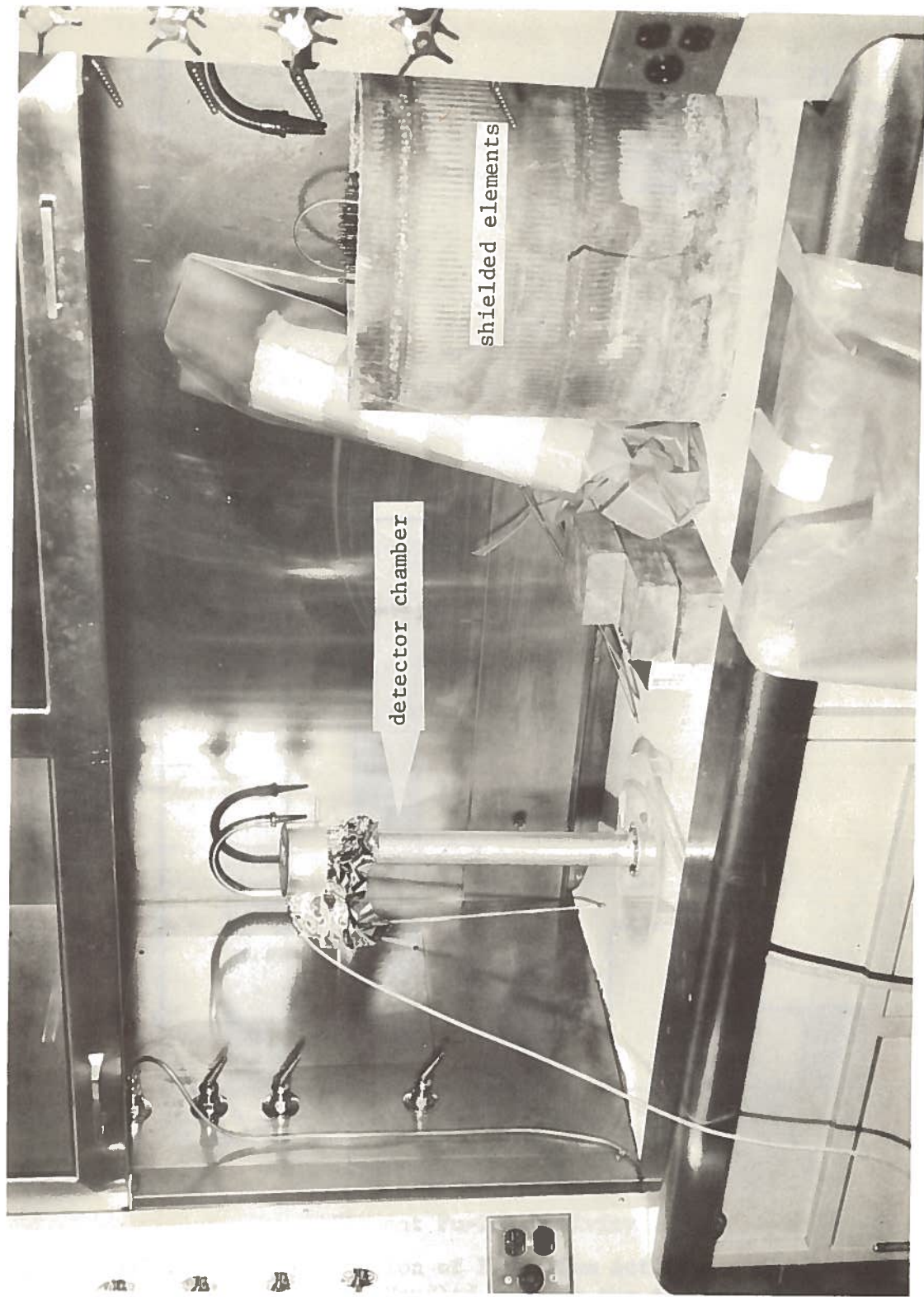


FIGURE 4. Setup for Counting Contaminated Elements.

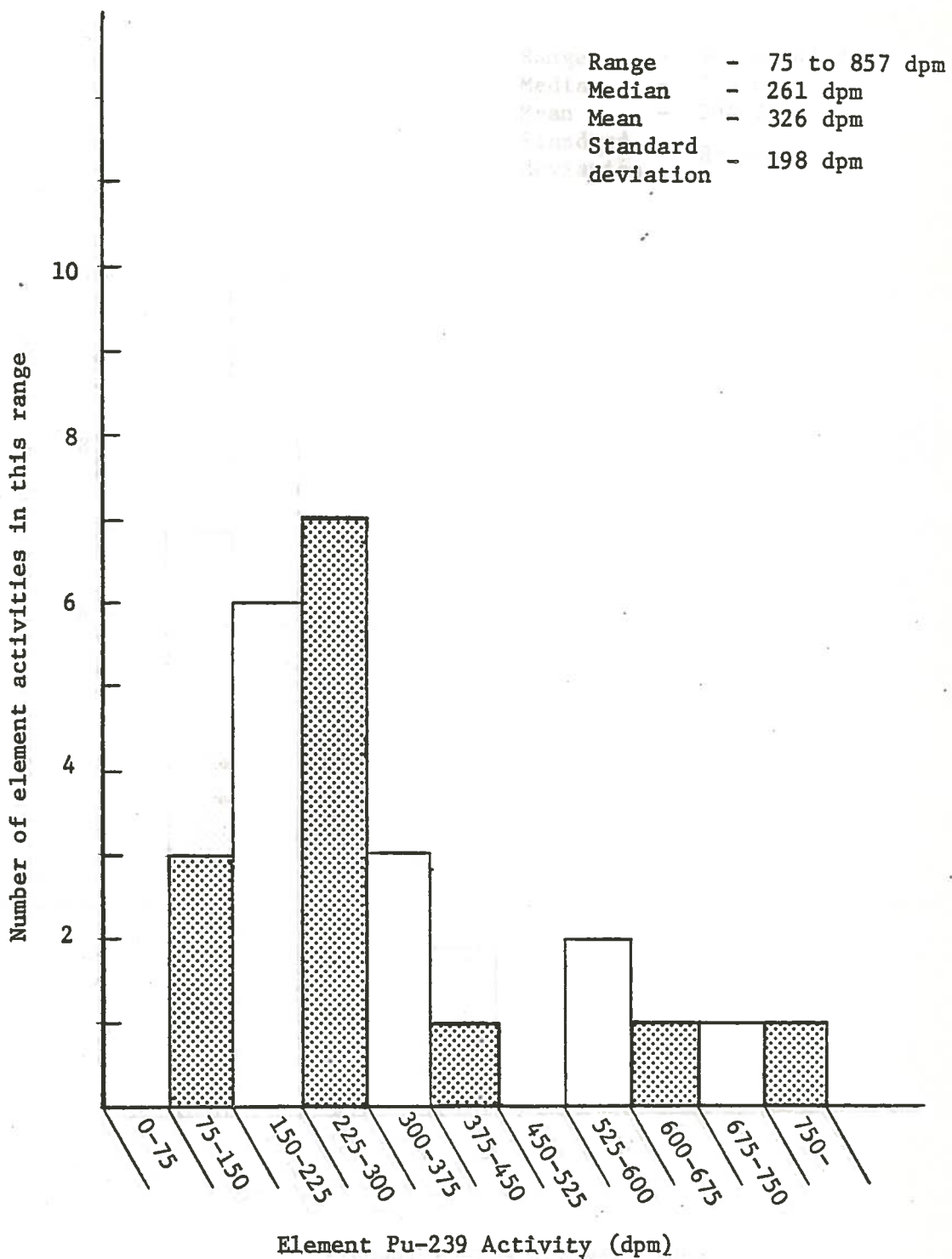


FIGURE 5. Distribution of Plutonium Activities for Elements Handled Only in the Air Cell (Elements 1-25).

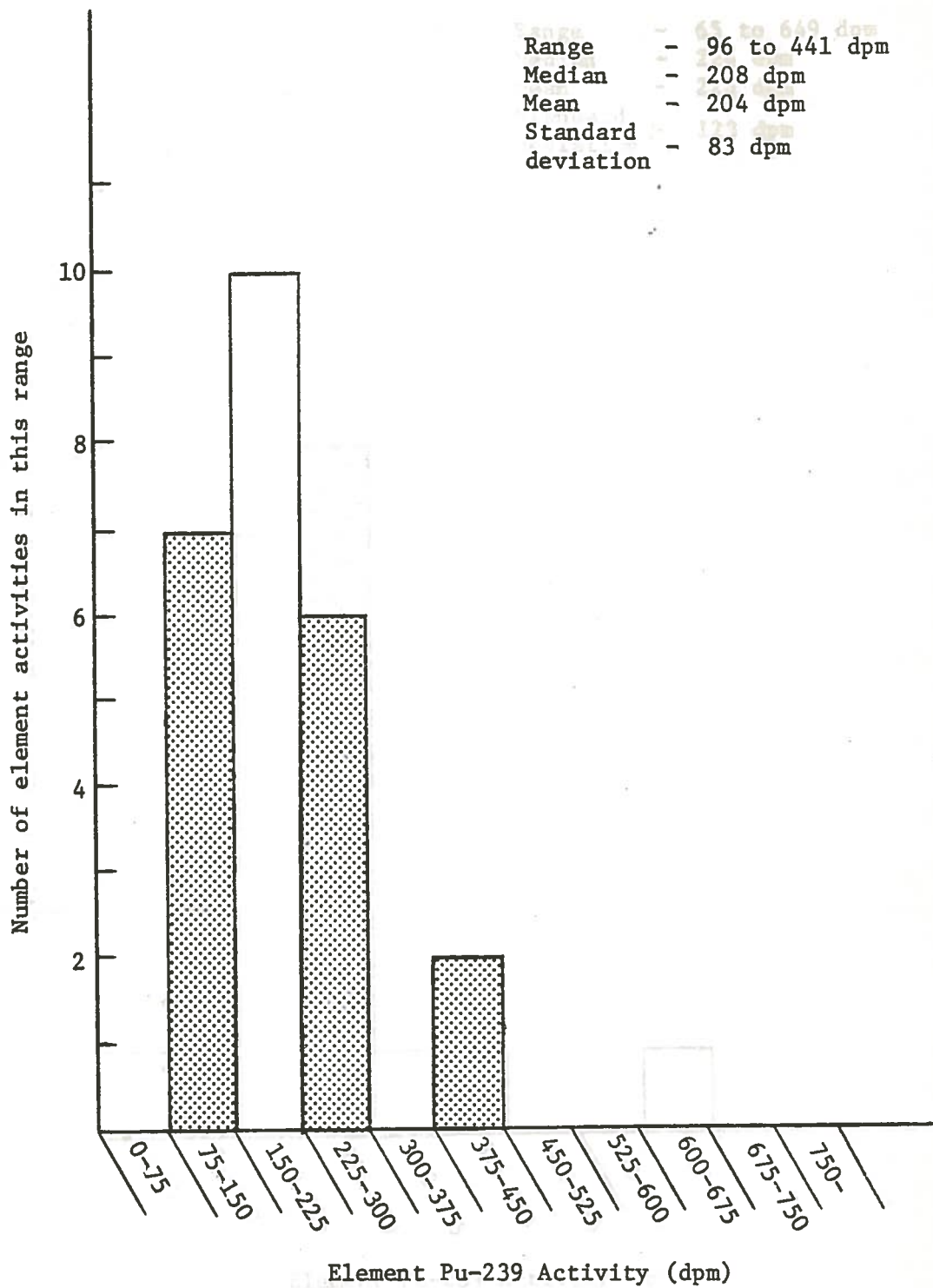


FIGURE 6. Distribution of Plutonium Activities for Elements Stored in the Argon Cell for Two Months (Elements 26-50).

As previously mentioned, the alpha counter was used to measure the activity of the elements.

ground level.

Table IV illustrates the results of the measurements.

Incidentally,

Range - 65 to 649 dpm
Median - 224 dpm
Mean - 225 dpm
Standard deviation - 123 dpm

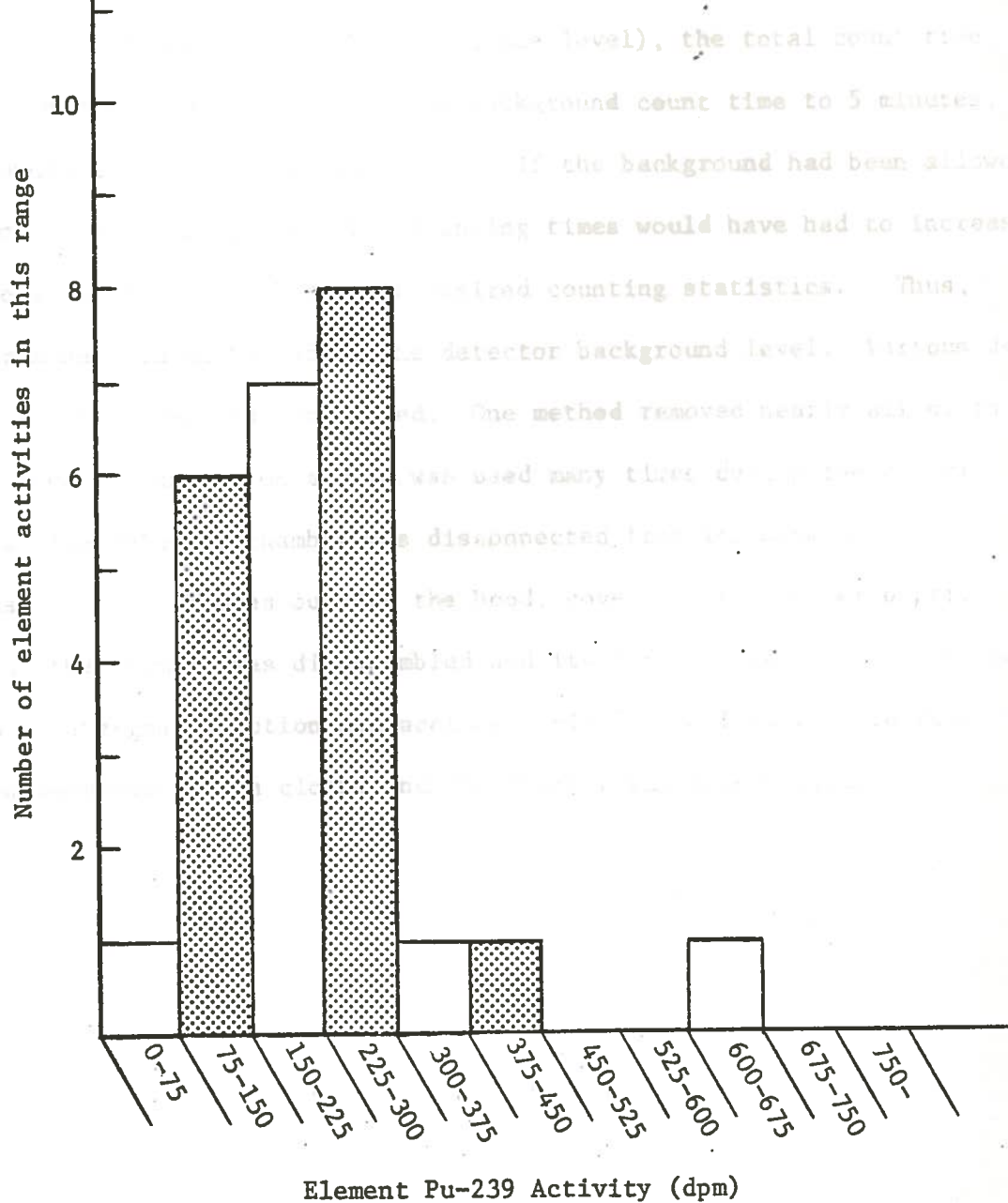


FIGURE 7. Distribution of Plutonium Activities for Elements Stored in the Argon Cell for Four Months (Elements 51-75).

As more elements were counted in the alpha counter, detector background levels increased considerably. Table IV illustrates this gradual increase in the background as elements 1 to 25 were counted.

The principal effect of the increased background was a reduction in the counting accuracy. As an accuracy of at least 10% was desired for all element counts (for a 95% confidence level), the total count time was extended to 30 minutes and the background count time to 5 minutes, in counting elements 23, 24, and 25. If the background had been allowed to increase still further, the counting times would have had to increase as well in order to achieve the desired counting statistics. Thus, efforts were taken to reduce the detector background level. Various decontamination methods were tried. One method removed nearly all of the deposited contamination and so was used many times during the experiment. The detector chamber was disconnected from its base and transferred to an area outside the hood, covered with blotter paper. Here, the chamber was disassembled and its inner surfaces were scrubbed with a detergent solution and acetone. Finally, all parts were rubbed dry with clean cotton cloths and the chamber was reassembled.

TABLE IV in the Primary

THE INCREASE IN THE DETECTOR BACKGROUND LEVEL AS ELEMENTS 1 TO 25 WERE COUNTED

Background Count Taken After Element Number	2-Minute Background Count
1	7
4	10
6	6
10	15
12	19
15	16
19	34
23	84
25	84

... the reactor primary tank ...
 ... loading Argon Gas ...
 ... sodium flow will be required ...
 ... operation. Thus, ...
 ... loading argon gas in the ...

Conditions in the Transfer Devices and in the Primary Tank

The preceding investigations determined how much plutonium and uranium is typically carried out-of-cell by stainless steel specimens but not how much is carried over and left behind in EBR-II.

Many fuel elements, after having been examined in HFEF/S, are reconstituted into experiment subassemblies and returned to EBR-II. Subassemblies are transferred to EBR-II from HFEF/S by way of the interbuilding transfer coffin (IBC) and the fuel unloading machine (FUM). While in these devices, hot argon gas is circulated through a subassembly at a flow rate of 60 standard cubic feet per minute and at a temperature of 410°F. Once in the primary tank, the subassembly is immersed in liquid sodium. If the subassembly is placed in the core or blanket region of the reactor, sodium flow will be experienced whenever the primary pumps are in operation. Thus, surface alpha contamination may be removed by hot flowing argon gas in the transfer devices or by molten sodium in the reactor primary tank.

Removal by Hot Flowing Argon Gas

Test elements for the study were contaminated dummy fuel elements described in a previous section. Each element was counted for plutonium before and after hot argon gas was blown across its surface.

Gas temperature and flow rate were selected to approximate the parameters for argon circulating through the IBC and the FUM, where argon gas is preheated to a temperature of about 410°F and circulated at a rate of 60 standard cubic feet per minute through a subassembly. Assuming that gas flows at an equal rate across each of the 91 fuel

elements in a driver fuel subassembly, then the flow rate of gas across each element is slightly less than 40 cubic feet per hour.

The test setup is shown in Figure 8. The setup basically consisted of a gas preheater coil which provided a bulk temperature increase in the gas, a nichrome wire resistance heater which provided rapid changes in the gas temperature, and a gas flow chamber, into which contaminated elements were inserted (equipment making up the gas heating system and gas flow chamber is listed in Table V). The gas flow through the chamber was set at 35 cubic feet per hour (cfh) to approximate the flow rate across elements in a subassembly. The gas temperature, monitored by a chromel-alumel thermocouple, was adjusted to 410°F.

The test was conducted on two sets of contaminated elements; ten elements from numbers 1 to 25 and five elements from numbers 26 to 50. Of the first 10 elements, five were subjected to flow for 5 minutes, and five for 30 minutes. All five elements of the second test set were subjected to a 30-minute argon flow.

Tables VI and VII illustrate the experiment results. The average ratio of alpha activities before and after argon flow were 0.86, 0.85, respectively, for the 5- and 30-minute flow sets in the first test. An average ratio of 0.86 was observed for elements of the second test set. Thus, contamination removal was not dependent on gas flow time.

There is evidence to believe that most of this reduction in alpha activity was due more to element handling than to hot flowing argon gas. Table VIII shows a comparison of the alpha activities of eight elements

before and immediately after the elements were subjected to a routine sequence of handling. Each element was transferred from its carrying grid slot to the alpha counter chamber, returned to the carrying grid, and then reloaded into the detector chamber. The data show that this sequence of handling removed about 25% of the surface contamination. Handling in the gas flow experiment was the same as this sequence except that elements were transferred to the experiment flow chamber instead of to the carrying grid. Furthermore, there was much more contact between the flow chamber inner walls and element surfaces than between elements and carrying grid. Thus, it is highly probable that the 15% reduction in the element surface contamination was due more to handling than to flowing gas.

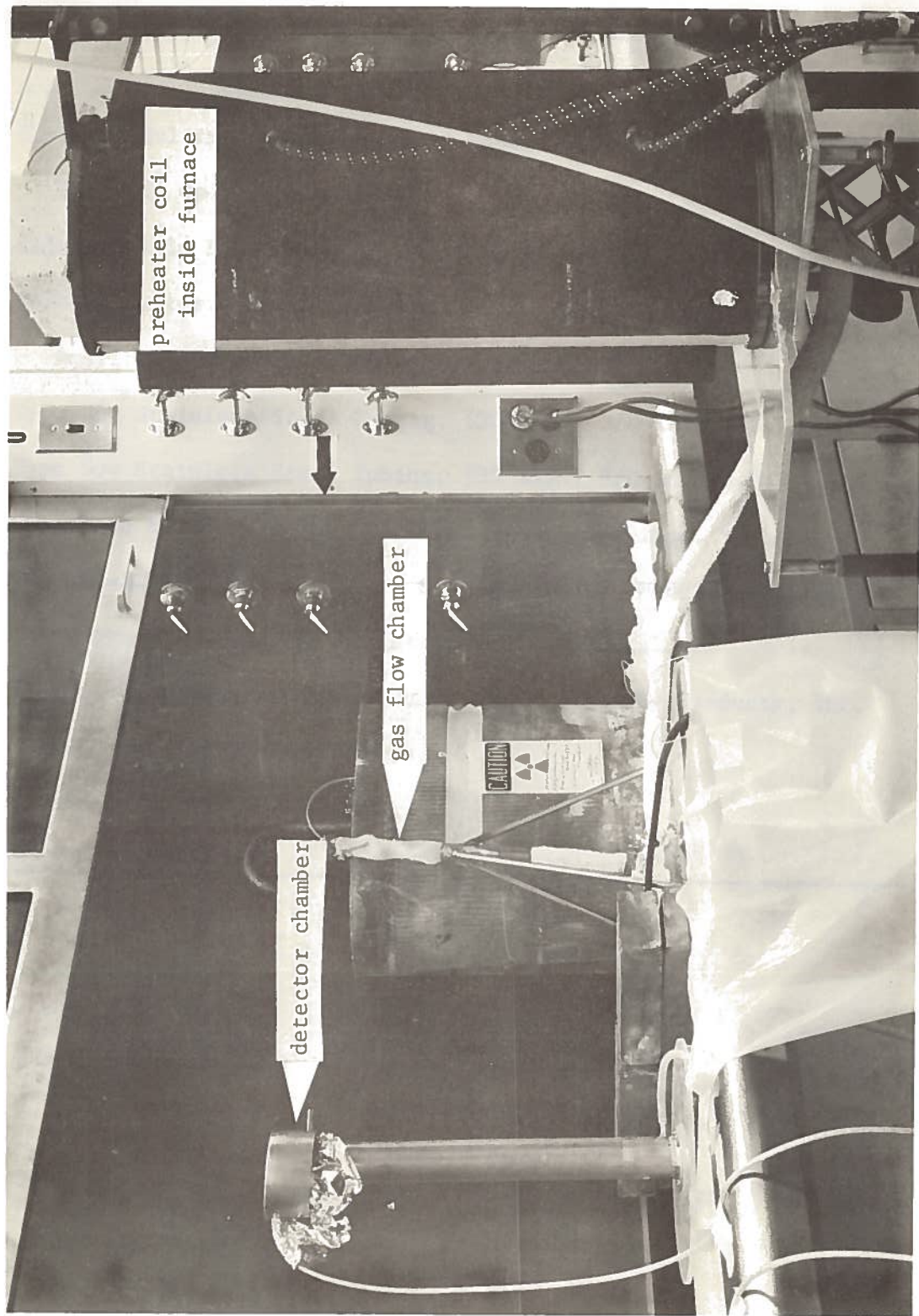


FIGURE 8. Hot Argon Gas Flow Experiment Setup.

After
Before

0.94

0.84

0.88

0.79

0.87

0.86

0.90

0.89

0.89

0.88

0.93

0.83

TABLE V

EQUIPMENT USED IN GAS FLOW EXPERIMENT SETUP

Electric Multiple Unit Furnace. Model Number M-2024S. Heavy duty
Electric Co. 2400 watts.

Adjust-A-Volt Power Supply. Type 500B. Staco Co. 0 to 140 volts.

Aerorod Heater. Model Number BX-A09. AERO Research Institute of
American Standards. 500 watts maximum output.

Type 304 Stainless Steel Tubing, (20 mils) 3/8-inch O.D.

Type 304 Stainless Steel Tubing, (35 mils) 3/8-inch O.D.

Stainless Steel Swagelok Fittings, 3/8-by-3/8-inch.

Chromel-Alumel Thermocouple. Size 24 A.W.G. Model Number 9B2C7.

Honeywell. External Resistance 10 ohms total.

Thermocouple Meter. Model Number 461. Assembly Products, Inc.

External Resistance 10 ohms.

Asbestos Sheath Around Tubing.

TABLE VI
 A COMPARISON OF ELEMENT ALPHA ACTIVITIES BEFORE AND
 AFTER HOT ARGON FLOW - FIRST TEST SET*

Element Number	Argon Flow Time (minutes)	Net Activity Before (cpm)	Net Activity After (cpm)	Ratio: $\frac{\text{After}}{\text{Before}}$
2	5	142	133	0.94
10	5	125	105	0.84
24	5	235	207	0.88
22	5	57	45	0.79
23	5	135	118	0.87
Average				0.86
24	30	199	179	0.90
13	30	112	100	0.89
17	30	310	213	0.69
21	30	232	199	0.86
20	30	222	207	0.93
Average				0.85

*Argon temperature = 410°F, flow rate = 35 cfh/

TABLE VII
 A COMPARISON OF ELEMENT ALPHA ACTIVITIES BEFORE AND
 AFTER ARGON FLOW - SECOND TEST SET*

Element Number	Net Activity Before (cpm)	Net Activity After (cpm)	Ratio: $\frac{\text{After}}{\text{Before}}$	Ratio: $\frac{\text{After}}{\text{Before}}$
39	99	99	1.00	0.78
40	119	97	0.81	0.93
43	122	108	0.88	0.87
46	216	159	0.74	0.72
48	109	95	0.87	0.79
			Average 0.86	0.68
				Average 0.47
				0.58

*Argon temperature = 410°F, flow rate = 35 cfh, flow time = 30 minutes.

TABLE VIII
 A COMPARISON OF ELEMENT ALPHA ACTIVITIES BEFORE
 AND AFTER HANDLING

Element Number	Net Alpha Activity Before (cpm)	Net Alpha Activity After (cpm)	Ratio: $\frac{\text{After}}{\text{Before}}$
2	181	142	0.78
10	134	125	0.93
13	129	112	0.87
17	428	310	0.72
20	280	222	0.79
21	341	232	0.68
22	122	57	0.47
24	406	235	0.58
		Average	<u>0.73</u>

Removal by Hot Molten Sodium

Five contaminated fuel elements were immersed in a stainless steel tube filled with about 100 cm³ of 700°F sodium (see Figure 9). The sodium level was adjusted to completely cover the elements. The tube was wrapped with a Briskeat resistance-heating tape, the output of which was controlled by a variac. Sodium temperature was monitored by a chromel-alumel thermocouple submerged in the sodium.

The experiment was carried out in a glovebox, with a pure argon atmosphere. Oxygen levels inside the glovebox were held to less than 10 ppm due to sodium's reactivity with oxygen.

The basic experiment procedure was the following:

- 1) Elements counted for alpha activity prior to sodium wash.
- 2) Elements transferred from hood to the glovebox and inserted into sodium wash tube.
- 3) Tube filled with sodium. Sodium temperature adjusted to 700°F.
- 4) Elements intermittently agitated in sodium for 3 hours.
- 5) Elements removed from wash tube and transferred out of glovebox.
- 6) Sodium removed from element surfaces.
- 7) Elements counted for alpha activity.
- 8) Removed sodium residue analyzed for plutonium.

The sodium residue on element surfaces was removed to prevent masking of alpha activity by the residue. Each element was immersed singly in an ethanol (95% ethyl alcohol, 5% water) solution. The

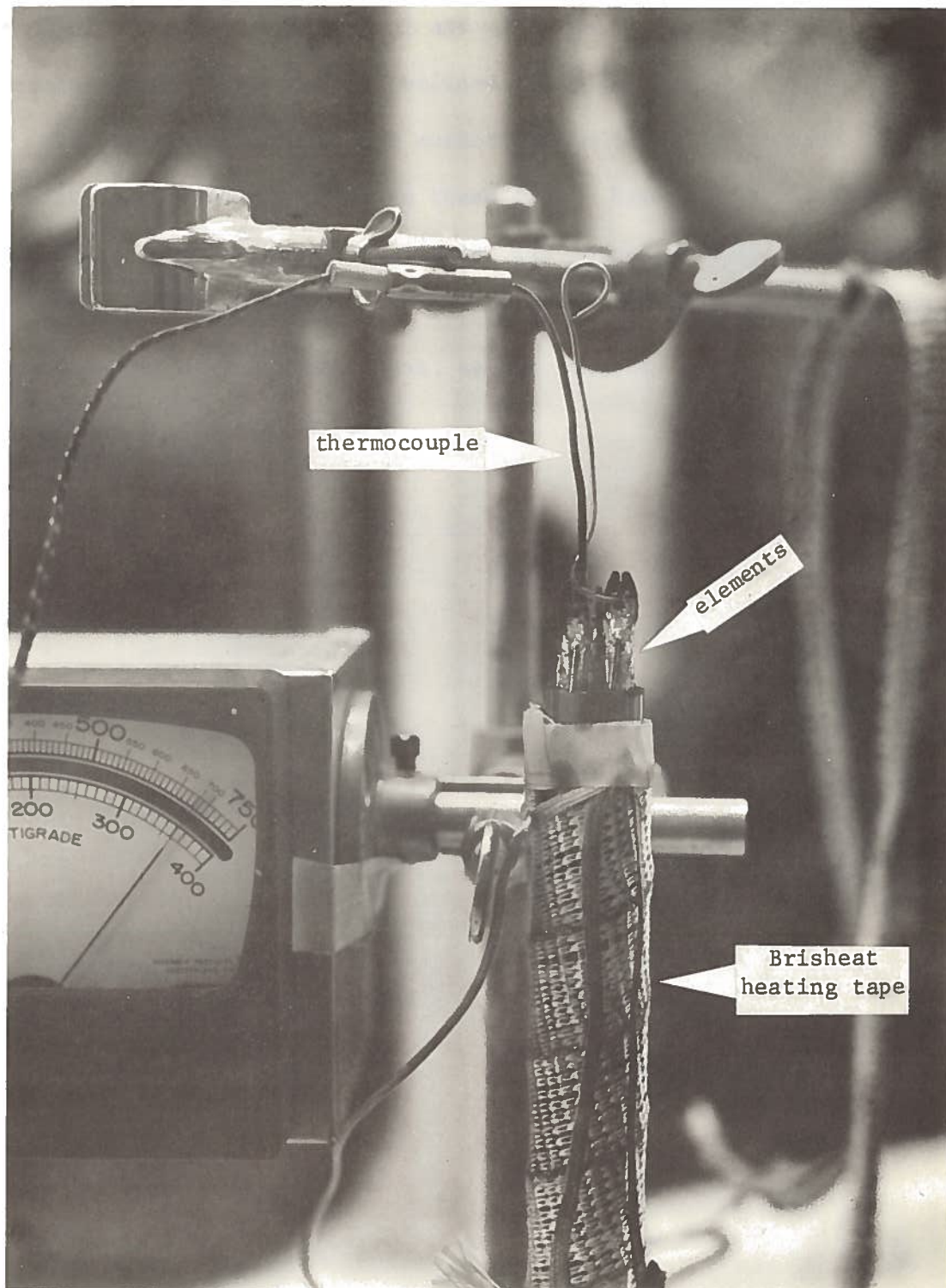


FIGURE 9. Contaminated Elements Immersed in 700°F Sodium.

elements were then rinsed with water, rinsed again with ethanol, and air dried. Finally, the ethanol and water washes were analyzed for plutonium using the procedure outlined in Appendix B.

The experiment results are summarized in Table IX. As can be seen, there is considerable scatter in these data. However, on the average, about 30% of the surface contamination was not removed by the experiment process. Furthermore, only about 10% of the surface activity was removed in the ethanol-water wash, so the majority of the contamination was removed in the sodium wash.

TABLE IX
RESULTS OF THE SODIUM WASH EXPERIMENT

Element Number	Net Alpha Activity Before Wash (dpm/element)	Net Alpha Activity After Wash (dpm/element)	Ratio of Final/Initial Activity	Activity Removed by Sodium Removal Process (dpm)	Fraction of Total Alpha Activity Removed by Sodium Removal Process
51	304	131	0.43	31	0.10
61	318	143	0.45		
65	304	88	0.29		
68	263	57	0.22		
75	616	55	0.09	51	0.08
			Average 0.30		

DISCUSSION

Summary of Results

The most significant results from the study were the following:

An alpha counter which was capable of monitoring plutonium on non-irradiated fuel element surfaces was designed and tested. It is feasible to assemble and load this detector chamber remotely. However, it cannot monitor alpha activities on elements having radiation levels greater than about 10 R/hr due to pulse pileup limitations.

Plutonium and uranium levels of 10^{-5} $\mu\text{g}/\text{cm}^2$ and less than 0.04 $\mu\text{g}/\text{cm}^2$, respectively, were observed on stainless steel fallout samples stored in-cell. There was no significant buildup of uranium and plutonium on block surfaces with time in-cell. Levels of the order of 10^{-3} $\mu\text{g}/\text{cm}^2$ plutonium and 1 $\mu\text{g}/\text{cm}^2$ uranium were observed on blocks handled in the cell. The maximum observed ratio of U/Pu contamination adhering to block surfaces was about 1000:1.

About 2.5×10^{-5} $\mu\text{g}/\text{cm}^2$ plutonium were observed on elements routinely handled and examined in the cell. The maximum level was about 10^{-4} $\mu\text{g}/\text{cm}^2$. There was no significant increase in the amount of contamination carried out-of-cell due to storage in the argon cell. On the contrary, stored elements were, statistically, less contaminated.

Less than 15% of the surface contamination was removed by argon gas, at approximately the same temperature and flow rate of argon

flowing through EBR-II fuel transfer devices. There is evidence to believe that most of this activity was removed by handling and not by hot flowing argon gas. The amount of contamination removed was not dependent on gas flow time.

On the average, about 40% of the contamination was not removed by molten 700°F sodium.

Discussion of Results

Contamination on nonirradiated fuel element surfaces could be accurately determined with an ionization chamber operated in the pulse mode. However, this counter could not be used with irradiated fuel elements because of the high background caused by beta and gamma pulse pileup. Several investigators have indicated that large backgrounds of beta and gamma radiation can produce pulses of sufficient size to pass the discriminator. ^(16,19) This would explain why the background pulse heights increased as argon flowed into the chamber, displacing the oxygen. Oxygen has a high affinity for electron attachment; ⁽¹⁶⁾ hence, most of the electrons freed by ionizations in the air and by photoemission from the detector walls would not have been collected at the anode. Argon has a small affinity for electron attachment at low gas pressures; so, most of the freed electrons would have been collected after oxygen was purged from the chamber. After argon flow had been terminated, oxygen could diffuse into the detector chamber, scavenging the freed electrons. This would serve to gradually decrease the background level.

Analysis of uranium and plutonium, chemically removed from twenty block surfaces led to only four explicit ratios of U/Pu contamination. Three were clustered between 200 and 300:1, while one ratio was about 900:1. Explicit ratios could not be determined for the remaining 16 blocks because of the extremely low levels of uranium, on their surfaces. Data in Tables II and III indicate that these contamination ratios could vary from 0 to 1000:1.

There are two possible explanations for the variance of explicit ratios: (1) an error was made in either the uranium or plutonium determination, or (2) a smaller amount of plutonium was intermixed with the uranium contamination. It is unlikely that an error was made in either of the chemical determinations. These methods have been routinely used for measuring microgram quantities of uranium and plutonium for years. The most likely cause for the high ratio would be that a smaller amount of plutonium was intermixed with the uranium. During the period in which HFEF/S was a fuel processing facility, fuels of varying degrees of burnup were refined in the argon cell (from about 0.1 atom % to about 2 atom %). It would, therefore, be expected that some fuel which was melt refined in HFEF/S had little plutonium inbred with the uranium. It is possible that some fuel particles, which contained little plutonium, were not collected in the refining process and were scattered randomly by the 60 miles-per-hour winds in the argon cell.

About 90% of the observed plutonium levels on typical fuel elements were clustered between 10^{-5} and 5.0×10^{-5} $\mu\text{g}/\text{cm}^2$. Elements

handled only in the air cell were statistically less contaminated than elements which were handled in the air cell and stored in the argon cell. Thus, if any additional contamination was picked up in storage, it was very loosely adherent for it was not carried out-of-cell.

The surface contamination has two parts: a loosely adherent and a strongly adherent part. The majority of the contamination is loosely adherent: about 50% to 90% is removed by a combination of molten sodium, ethanol, and handling. About 40% is not removed by molten sodium; so this contamination may not be removed by sodium in the primary tank. However, the contaminated elements were not subjected to flowing sodium. The 40% estimate of fixed contamination may, therefore, be too large. Finally, it is highly probable that no contamination is removed by hot argon gas, circulating through the transfer devices.

Estimates of Tramp Uranium and Plutonium Carried Over to EBR-II

Based on the experiments, the amount of fissionable material which remained adherent to experiment surfaces in the reactor core was estimated for a particular reactor loading configuration. The first step in making this calculation was to estimate the total contaminated experiment surface area in the core. The surfaces making up an experimental subassembly are the subassembly can surfaces and the fuel element surfaces. Fuel elements will become contaminated because they are handled in-cell; but, the outer can surfaces may also become contaminated. The outer can surfaces are handled considerably in the

reconstitution process. The inner can surfaces, however, are handled very little. Thus, it was assumed that contaminated experiment surfaces were the fuel element and the outer can surfaces.

Several different types of experiment subassemblies commute between HFEF/S and EBR-II, and the number of each type in the reactor varies from one reactor run to the next. Calculations of fixed and removable activity were based on the reactor loading configuration for Run 65B. Tables X and XI list parameters for the various experiment fuel elements that were in the reactor for this particular run. As can be seen in Table XI, the total experiment fuel element surface area in the core was about 32,000 cm². The total experiment outer can surface area in the core was about 10,000 cm². Therefore, the total contaminated experiment surface area in the core was about 42,000 cm².

The next step was to estimate how much uranium and plutonium contamination was carried out-of-cell by subassemblies. From the experiment results, the maximum plutonium level carried out-of-cell by fuel elements was about 10⁻⁴ μg/cm². The maximum ratio of U/Pu contamination in-cell was about 1000:1; thus, the maximum amount of uranium carried out-of-cell was about 0.1 μg/cm².

The final step was to estimate what fraction of the contamination was removed in the transfer devices and in the primary tank. Based on results of the contamination adherence experiments, it was assumed that none of the contamination was removed in the IBC or FUM and that about 60% of the contamination was removed in the primary sodium.

TABLE X
EXPERIMENT ELEMENTS OR CAPSULES IN REACTOR FOR RUN 65B*

Subassembly Type	Number of Capsules in Subassembly	Capsule Diameter (inches)	Capsule Length (inches)	Spacer Wire Diameter (inches)	Total Outside Capsule Plus Spacer Wire Surface Area per Subassembly (cm ²)	Total Outside Capsule Plus Spacer Wire Surface Area in Core per Subassembly (cm ²)
A19A	19	0.375	40.0	0	5.8 x 10 ³	2.0 x 10 ³
B7A	7	0.805	57.9	0	6.6 x 10 ³	1.6 x 10 ³
B7C	7	0.805	57.9	0	6.6 x 10 ³	not in core
E19D	19	0.250	60.9	0.062	7.3 x 10 ³	1.7 x 10 ³
E37C	37	0.230	60.9	0.040	12.3 x 10 ³	2.9 x 10 ³
E37D	37	0.230	60.9	0.040	12.3 x 10 ³	2.9 x 10 ³
F37A	37	0.250	60.9	0.062	14.2 x 10 ³	3.3 x 10 ³
F37B	37	0.250	60.9	0.062	14.2 x 10 ³	not in core
H37A	37	0.250	60.9	0.040	13.2 x 10 ³	3.1 x 10 ³
J37B	37	0.230	40.0	0.056	8.6 x 10 ³	3.0 x 10 ³
Q19A	19	0.230	40.0	0.056	4.4 x 10 ³	1.6 x 10 ³
T19	19	0.290	60.9	0.056	8.1 x 10 ³	not in core
M1A	1	2.18	40.0	0	1.8 x 10 ³	0.63 x 10 ³

*Handled in HFEF/S.

TABLE XI
TOTAL STAINLESS STEEL SURFACE AREA OF EXPERIMENT
ELEMENTS OR CAPSULES IN REACTOR FOR RUN 65B*

Subassembly Type	Number of Sub-assemblies of This Type in EBR-II Tank	Number of Sub-assemblies of This Type in EBR-II Core	Total Outside Surface Area of All Capsules of This Type in		Total Outside Surface Area of All Capsules of This Type in EBR-II Core (cm ²)
			Tank (cm ²)	SS (cm ²)	
A19A	4	3	23.2 x 10 ³	6.1 x 10 ³	6.1 x 10 ³
B7A	3	2	19.8 x 10 ³	3.2 x 10 ³	3.2 x 10 ³
B7C	1	0	6.6 x 10 ³	-----	-----
E19D	1	1	7.3 x 10 ³	1.7 x 10 ³	1.7 x 10 ³
E37C	1	1	12.3 x 10 ³	2.9 x 10 ³	2.9 x 10 ³
E37D	1	1	12.3 x 10 ³	2.9 x 10 ³	2.9 x 10 ³
F37A	4	2	56.8 x 10 ³	6.6 x 10 ³	6.6 x 10 ³
F37B	1	0	14.2 x 10 ³	-----	-----
H37A	1	1	13.2 x 10 ³	3.1 x 10 ³	3.1 x 10 ³
J37B	1	1	8.6 x 10 ³	3.0 x 10 ³	3.0 x 10 ³
Q19A	1	1	4.4 x 10 ³	1.6 x 10 ³	1.6 x 10 ³
T19	1	0	8.1 x 10 ³	-----	-----
MLA	1	1	1.8 x 10 ³	0.63 x 10 ³	0.63 x 10 ³
	<u>21</u>	<u>14</u>	<u>188.6 x 10³ cm² SS</u>	<u>31.7 x 10³ cm² SS</u>	<u>31.7 x 10³ cm² SS</u>

*Handled in HFEF/S

Combining together all of the above, the maximum amount of plutonium and uranium which remained on experiment surfaces in the core for Run 65B were:

$$\begin{aligned} \text{Maximum amount of plutonium} &= (0.4)(10^{-4} \mu\text{g}/\text{cm}^2)(42 \times 10^3 \text{ cm}^2) \\ \text{remaining on experiment sur-} & \\ \text{faces in the core} &= 1.7 \mu\text{g} \end{aligned}$$

$$\begin{aligned} \text{Maximum amount of uranium} &= (1000)(1.7 \mu\text{g}) \\ \text{remaining on experiment sur-} & \\ \text{faces in the core} &= 1.7 \text{ mg} \end{aligned}$$

It can be assumed that all of the plutonium was Pu-239. If it is assumed that half of the uranium was U-235 and half was U-238, a maximum of 0.9 mg U-235 and 0.9 mg U-238 remained on surfaces in the core. This assumption is justified because the composition of typical fuel processed in HFEF/S was about 45% U-235, 50% U-238, and 5% fission (Zirconium, Molybdenum, Ruthenium, and Palladium).⁽¹⁴⁾ As the fission rate for U-238 in the core region is only about 2 - 3% of the U-235 fission rate,⁽⁴⁾ the above calculation implies that there was the equivalent of about 0.9 mg tramp U-235 on experiment surfaces in the core. This is comparable to Brunson's estimate of the equivalent of 2 - 3 mg of tramp U-235 in the reactor core.⁽⁴⁾

It is also interesting to estimate how much uranium and plutonium was washed off of all reconstituted experiment subassemblies which were in the primary tank for Run 65B. From Table XI, the total experiment fuel element surface area in the reactor for Run 65B was about 190,000 cm². The total outer can surface area was about 100,000.

Hence, the total contaminated experiment surface area in the reactor was about 290,000. The maximum amounts of uranium and plutonium washed off of these 21 experimental subassemblies were:

$$\begin{aligned} \text{Maximum amount of} &= (0.6)(10^{-4} \mu\text{g}/\text{cm}^2)(2.90 \times 10^5 \text{ cm}^2) \\ \text{plutonium washed off} &= 17 \mu\text{g} \end{aligned}$$

$$\begin{aligned} \text{Maximum amount of} &= (1000)(17 \mu\text{g}) \\ \text{uranium washed off} &= 17 \text{ mg} \end{aligned}$$

About 150 experiment subassemblies and about 400 driver fuel subassemblies, fabricated in HFEF/S, have gone over to EBR-II. If it is assumed that proportionate amounts of contamination were washed off of all 550 subassemblies, then the total amount of uranium and plutonium which was washed off were:

$$\begin{aligned} \text{Total plutonium} &= \frac{(550)(17 \mu\text{g})}{(21)} \\ \text{washed off} &= 0.5 \text{ mg} \end{aligned}$$

$$\begin{aligned} \text{Total uranium} &= (1000)(0.5 \text{ mg}) \\ \text{washed off} &= 500 \text{ mg} \end{aligned}$$

The most recent analyses indicated that there were less than 2 ppb uranium and less than 0.004 ppb plutonium in the primary sodium. (5) This implies that there should be less than 1.2 mg plutonium and less than 600 mg uranium homogeneously distributed in the primary tank sodium. Thus, the above estimates agree with what is currently observed.

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

Based on the results and calculations of this study, five principal conclusions were drawn:

- 1) Less than 10^{-4} $\mu\text{g}/\text{cm}^2$ plutonium and less than $0.1 \mu\text{g}/\text{cm}^2$ uranium are presently being carried out of HFEF/S by experiments.
- 2) Virtually none of the contamination picked up in-cell is removed by hot argon gas, circulating through the transfer devices.
- 3) Less than 40% of the contamination typically remains on experiment surfaces immersed in the primary tank sodium.
- 4) Less than $1.7 \mu\text{g}$ Pu-239 and less than 0.9 mg U-235 remain on experiment surfaces in the core. Fixed Pu-239 contamination is not the source of the background fission product gas activity in the reactor cover gas. However, fixed U-235 contamination may be the source of the cover gas activity.
- 5) The alpha counter, utilized in this study, cannot be used to monitor plutonium levels on nonirradiated elements.

The estimate of 0.9 mg U-235 contamination, remaining on experiment surfaces in the reactor core, is comparable to Brunson's estimate of the equivalent of $2 - 3 \text{ mg}$ of tramp U-235 in the core. ⁽⁴⁾

However, actual fixed amounts would be considerably less if: (1) typical levels of uranium carried out-of-cell are less than $0.1 \mu\text{g}/\text{cm}^2$, and (2) if more than 60% of the contamination is washed off by flowing sodium in the primary tank. If this were the case, uranium contamination, remaining on experiment surfaces in the core, would not be the principal source of the background fission product gas activity in the reactor cover gas.

Estimates of the total amounts of plutonium and uranium deposited in the primary sodium agree with what is currently observed. The amounts suspended in the primary sodium would be much less if some of the contamination plated out on primary tank components.

Recommendations

This study has determined how much plutonium and uranium is typically carried to EBR-II by reconstituted experiments. However, it has not adequately determined how much of this contamination is deposited in the EBR-II primary tank. The sodium wash experiment indicated that most of the contamination on element surfaces is removed by 700°F stagnant sodium, but it did not determine how much is typically removed by flowing sodium in the primary tank.

To study this problem, it is recommended that an experimental subassembly, containing nonirradiated elements, be sent to the storage basket of EBR-II. Plutonium levels could be monitored with the alpha counter described in this study because elements would not be irradiated in the storage basket. Uranium contamination levels could

be estimated from the approximate ratio of U/Pu in-cell. The results of this experiment would yield a more quantitative estimate of how much contamination is removed from experimental subassemblies by sodium flowing through the reactor.

In addition to the above, three tasks should be initiated after the onset of destructive examinations in HFEF/N:

- 1) Determine how much plutonium and uranium is typically carried out of HFEF/N by fuel elements. This study would determine whether contamination levels on experiments were significantly increasing. The alpha counter, described in this study, could again be used to monitor plutonium levels on element surfaces. Corresponding uranium levels could again be estimated from the approximate U/Pu ratio in HFEF/N.
- 2) Closely follow conditions in the reactor primary tank. This task, continued as long as EBR-II is in operation, would show if plutonium and uranium levels in the primary tank increase with time. This program would include: (a) analyzing the primary sodium at regular intervals for plutonium and uranium content, (b) smearing all components removed from the primary tank and analyzing them for plutonium and uranium, and (c) analyzing the cover gas at regular intervals for increases in the background fission product gas activity.
- 3) Conduct a survey of possible decontamination methods. This study would determine the feasibility of various decontamination methods. The ideal situation would be if contamination

levels on experiments leaving HFEF/N could be held near or less than currently observed levels. Included in this survey should be two very promising methods: a water-detergent scrub, and cleaning by ultrasonics. Only specimens handled and examined in HFEF/N should be used in this study.

... from High-Resolution ...
... American Nuclear Society

... Glenn Brunson, physicist Y24-11
... Division.

... with S. A. Haiker ...
... Y24-11

... with J. H. Cook ...
... Section.

... Fundamentals of Metallurgy ...
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REFERENCES

1. L. J. Koch, et al., Hazard Summary Report Experimental Breeder Reactor-II (EBR-II), (Argonne National Laboratory, Lemont, Illinois, 1957).
2. Private communication with Earl Ebersole, Analytical Laboratory Manager, EBR-II site.
3. G. S. Brunson, "Preliminary Results from High-Resolution Gamma Analysis of EBR-II Cover Gas," American Nuclear Society Transactions, Oct. 1971.
4. Private communication with Glenn Brunson, physicist, EBR-II Project, Analysis and Physics Division.
5. Private communication with S. A. Barker and R. Villarreal, chemists, Analytical Section, EBR-II site.
6. Private communication with J. H. Cook, engineer, HFEF/S, Experiment Coordination Section.
7. H. F. Henry, Fundamentals of Radiation Protection, p. 259 (Interscience, New York, 1969).
8. H. Kiefer and R. Maushart, Radiation Protection Measurement, p. 275 (Pergomon Press, New York, 1972).
9. H. Blatz, Radiation Hygiene Handbook, p. 22 (McGraw-Hill, New York, 1959).
10. R. A. Jaroszeski, et al., "Fluorometric Determination of Micro-quantities of Uranium in Mixtures of Uranium and Plutonium," Analytical Chemistry, 37, 766, 1965.

11. F. A. Centanni, et al., "Fluometric Determination of Uranium," *Analytical Chemistry*, 28, 1651, 1956.
12. Private communication with R. Villarreal.
13. G. S. Brunson and E. Ebersole, unpubl., 1973.
14. J. C. Hesson, et al., "Description and Proposed Operation of the Fuel Cycle Facility for the Second Experimental Breeder Reactor (EBR-II)," Argonne National Laboratory, 1963.
15. G. D. O'Kelley, Detection and Measurement of Nuclear Radiation, (National Academy of Science, Research Council, NAS-NS-3105, Oak Ridge, Tenn., 1962).
16. W. J. Price, Nuclear Radiation Detectors, (McGraw-Hill, New York, 1964).
17. I. Kaplan, Nuclear Physics, (Addison Wesley Publ. Co., Reading, 1955).
18. Fuel Handling and Examinations Capability, Argonne National Laboratory, 1971.
19. S. C. Curran, "The Proportional Counter as Detector and Spectrometer," *Handbuch der Physik*, Berlin, pp. 174-212, 1958.
20. F. S. Kirn, "EBR-II as a Fast Reactor Irradiation Facility," *Nuclear News*, 13, 62, 1970.
21. R. M. Adams, HFEF/N Hot Fuel Examination Facility/North Final Facility Safety Report, (Argonne National Laboratory, Argonne, 1972).

APPENDIX A

DESCRIPTION OF FACILITIES

Experimental Breeder Reactor-II (1,20)

Experimental Breeder Reactor-II (EBR-II) is an important facility for irradiations testing under the Atomic Energy Commission's Liquid Metal Fast Breeder Reactor Program. It is located in Southeastern Idaho on the National Reactor Testing Station and is operated by Argonne National Laboratory.

EBR-II is a sodium-cooled fast breeder reactor which utilizes enriched uranium as the driving fuel. At peak power the reactor has an output of 62.5 megawatts thermal and 20 megawatts electrical. The reactor essentially consists of three distinct regions: an enriched uranium fuel core, a stainless steel reflector region, and a depleted uranium blanket region. The blanket region breeds Pu-239 as a result of neutron capture.

The basic fuel and blanket unit is a hexagonal can, 2.3 inches across the flats, called a subassembly. All fuel and experimental elements are built into the same basic hexagonal can before they are transferred to EBR-II. Driver fuel subassemblies contain a bundle of 91 fuel elements. Each fuel element consists of a uranium metal pin 13.5 inches long, which is sodium bonded inside a stainless steel jacket of 0.174 inches O.D. (see Figure 10). A variety of experimental subassemblies are also run through the reactor. These subassemblies house a varying number of mixed-oxide or carbide

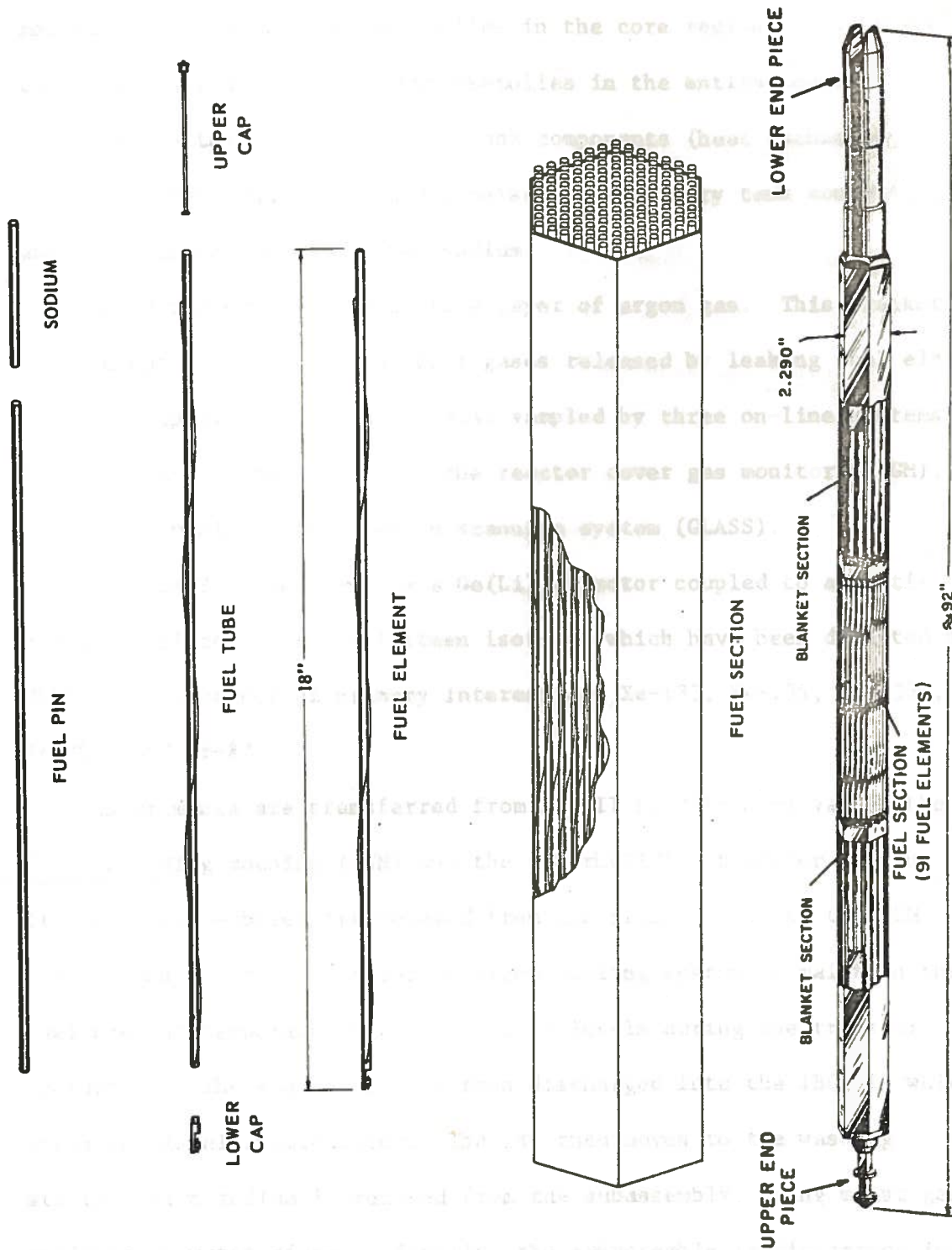


FIGURE 10. EBR-II Fuel Subassembly and Fuel Elements.

experiment elements or capsules. There can be as many as 91 experimental and driver fuel subassemblies in the core region, while there can be as many as 637 total subassemblies in the entire reactor.

The reactor and all primary tank components (heat exchanger, storage basket, and pumps) are immersed in a primary tank containing about 86,000 gallons of molten sodium.

Above the primary sodium is a layer of argon gas. This blanket is sampled daily for fission product gases released by leaking fuel elements or capsules. The gas is also sampled by three on-line systems: the fission gas monitor (FGM), the reactor cover gas monitor (RCGM), and the germanium-lithium argon scanning system (GLASS).

The GLASS system employs a Ge(Li) detector coupled to a multi-channel analyzer. Of the fifteen isotopes which have been detected by GLASS, the isotopes of primary interest are Xe-133, Xe-135, Xe-135m, Kr-85m, and Kr-87.

Experiments are transferred from EBR-II to HFEF/S by way of the fuel unloading machine (FUM) and the interbuilding transfer coffin (IBC). Subassemblies are removed from the primary tank by the FUM. The FUM has a forced circulation argon cooling system to maintain the fuel element temperatures at acceptable levels during the transfer operations. The subassembly is then discharged into the IBC, in which argon gas is also circulated. The IBC then moves to the washing station where sodium is removed from the subassembly, using moist gas followed by water rinses. Finally, the subassembly is discharged into the air cell by way of the large transfer port. Reconstituted

experiments follow the same route back to EBR-II in reverse order, bypassing the wash station.

The Hot Fuel Examination Facility/South (14,18)

The Hot Fuel Examination Facility/South (HFEF/S) consists of two cells - an argon cell and an air cell, surrounded by an operating area and auxiliary laboratories (see Figure 11). Their purpose is to provide a radiation shielded area where subassemblies can be disassembled or rebuilt, and where experiments can be inspected and tested by remote methods.

The argon cell is constructed in the shape of a 16 regular-sided polygon with the air cell extending out from the west side. The walls of both the air and argon cells are made of 5-foot thick, high density concrete.

The atmosphere in the argon cell is highly purified argon gas. This noble gas is used to prevent excessive reactions of the cell atmosphere with exposed fuel or sodium. The atmosphere in the air cell is filtered air.

There are two major transfer ports out of the air cell to its surroundings. The small air cell sample port is a 12-inch diameter, horizontal pipe through the 5-foot thick north wall of the air cell. Objects having a radiation reading less than 100 mR/hr at or near their surface are permitted to be transferred out through this port. The large transfer port is located in the northwest corner of the air cell. This port consists of three plugs with 6-inch, 2-foot, and 6-foot diameters, respectively.

There are three transfer locks between the air and argon cells.
 Two small lock openings through the wall separating the cells and a
 large one between the argon cell into a transfer
 cell floor. The runway extends
 from the cell floor.

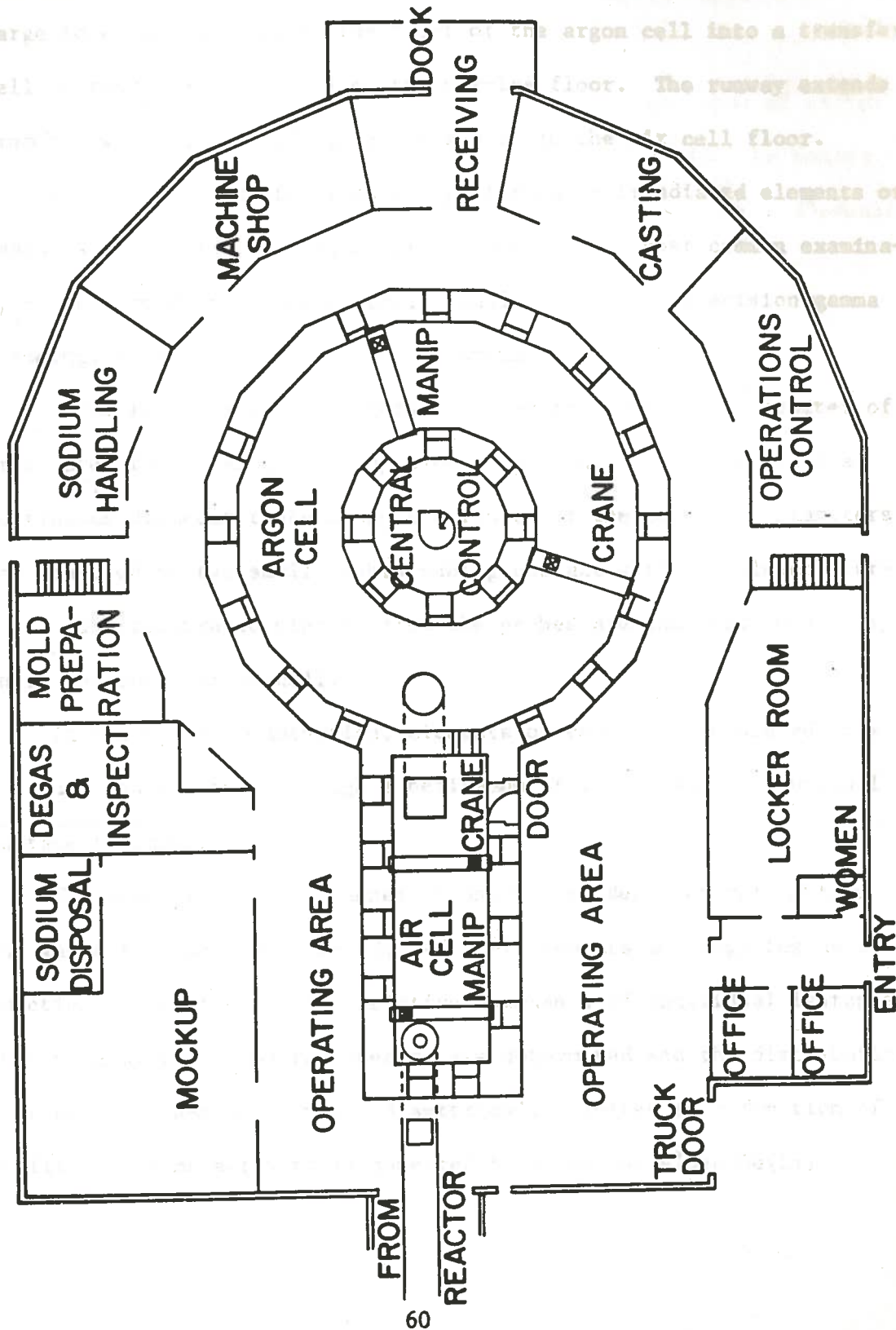


FIGURE 11. HFEF-S Main Floor Plan.

There are three transfer locks between the air and argon cells. Two small locks extend through the wall separating the cells and a large lock extends through the floor of the argon cell into a transfer cell or runway at the level of the service floor. The runway extends from below the argon cell to below a port in the air cell floor.

A variety of examinations are performed on irradiated elements or capsules in the air and argon cells. Some of the most common examinations performed are profilometry, visual inspection, precision gamma scanning, sodium bonding and bond testing.

A remote contact profilometer is used to measure the diameter of irradiated fuel elements or capsules. The profilometer provides a continuous diameter trace along the length of the capsule. Diameters are measured by two small probes making contact with the element surface. The calibrated signals from the probes are then fed to a strip chart recorder out-of-cell.

In the visual examination, elements or capsules are placed in a V-trough and examined through a periscope at close range for unusual surface features.

The precision gamma scanner is used to nondestructively obtain the gamma ray spectra of irradiated fuel elements and capsules as a function of position. The relative abundance of individual isotopes that make up the gamma-ray spectra are determined and the distribution of these isotopes axially and diametrically plotted as a function of position. Gamma activity is detected by a lead-shielded Ge(Li)

detector. Elements are placed behind a variable width collimator. Elements are positioned and collimator width varied, depending on the information desired.

The sodium bonder removes gas pockets from the layer of sodium between the irradiated fuel pin and the jacket. Inside the bonder, elements are heated to 450°C and then vertically impacted. Elements are then placed in the bond tester which is an eddy current instrument, used to detect voids in the sodium bond.

The Hot Fuel Examination Facility/North (21)

The Hot Fuel Examination Facility/North (HFEF/N) consists of two rectangular cells, the main cell and the decon cell, surrounded by an operating area and auxiliary laboratories (see Figure 12). The walls in both cells at the operating level are made of 4-foot thick, high density concrete.

The heart of HFEF/N is the main cell. Most experimental testing and examinations will be carried out in the pure argon atmosphere of this facility. Both nondestructive and destructive examinations will be remotely performed on mixed-oxide fuel elements, capsules and experiments. Many of the nondestructive examinations will be similar to examinations, presently performed in HFEF/S, such as profilometry, gamma-scanning and visual examinations. However, unlike HFEF/S, many plutonium-bearing fuel elements and capsules will be destructively examined and tested in the main cell. Elements and capsules will be cut, ground, polished, and etched. Large and small safety

test loops, containing ruptured fuel elements, will be dismantled and prepared for examination in the main cell. In all, the total amounts of contained and uncontained plutonium to be handled in the main cell will greatly exceed amounts handled in HFEF/S.

The main purpose of the decon cell will be to prevent the release of particulate contamination outside the hot cell environment. The decon cell is divided into two principal areas: the clean area and the decontamination area. The atmosphere in both cells will be filtered air. The decontamination area will be used primarily to prepare contaminated materials for entry into the clean area. Equipment being removed to repair areas for contact maintenance, will also be partially decontaminated in this cell. Some decontamination methods which may be used include dry vacuuming, air-jetting, washing with low and high pressure water sprays, and ultrasonic cleaning. The clean area will be used for packaging of experiments or waste materials for shipment off-site or to disposal facilities. This area will be kept, essentially, clean of radioactive contaminants.

There are two transfer locks between the main cell and the decontamination cell. The transfer tunnel and transfer lock will be used for moving large components between the main and decon cells. A small equipment transfer lock will be used to transfer small items between the two cells.

Experiments, reconstituted in HFEF/N, will be transferred to EBR-II in the following manner. Experiment subassemblies, housed

inside an IBC, will exit the main cell through the cask lock and into the cask tunnel. The IBC will then travel down this tunnel and exit HFEF/N through the truck lock. A truck will transport the IBC over to the interbuilding passageway in HFEF/S. From here, the transfer will follow a direct route between HFEF/S and EBR-II.

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Counting Procedure

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APPENDIX B

PROCEDURE FOR CHEMICALLY DETERMINING AMOUNTS OF PLUTONIUM AND URANIUM ON STAINLESS STEEL BLOCK SURFACES ⁽¹²⁾

This procedure outlines the steps in determining chemically quantities of uranium and plutonium on the stainless steel block surfaces. It is not an explicit procedure for making these determinations. It does, however, show the basic steps in the analytical processes. The lower detection limit for the plutonium method was 1 dpm/sample counted; the lower detection limit for the uranium method was 0.01 $\mu\text{g/ml}$ solution analyzed. ⁽²⁾

I. Plutonium Separation and Counting Procedure

- A. Place block in 100-ml beaker.
- B. Add 16 N HNO_3 concentrated.
- C. Heat to near boiling.
- D. Remove beaker from heat. Add 0.1 ml HF.
- E. Heat block in the above solution between 60 and 80°C for 5 to 10 minutes.
- F. Remove block, rinsing with H_2O . Reheat remaining HNO_3 - HF solution evaporating all but about 5 ml.
- G. Of this 5 ml solution, remove an aliquot for plutonium analysis; remaining amount is to be analyzed for uranium.
- H. Pour aliquot solution into separatory funnel.

- I. Add ascorbic acid.
- J. Add ammonium tartrate solution.
- K. Add thio - glycollic acid (mercaptoacetic acid).
- L. Make solution basic by adding ammonium carbonate.
- M. Add potassium cyanide.
- N. Add N-benzoyl-N-phenylhydroxylamine (BPHA) to solution.
- O. Add benzene to the separatory funnel. Mix up the aqueous solution with the benzene solution.
- P. Add 0.5N nitric acid to benzene solution.
- Q. Drain off the Pu-nitric acid solution into a stainless steel planchet.
- R. Evaporate the liquid. Alpha count the residue with a gas flow proportional counter.

II. Uranium Separation and Gross Uranium Counting

- A. Place nitric acid solution set aside for the uranium analysis into centrifuge cone.
- B. Add aluminum nitrate nonahydrate ($\text{Al}(\text{NO}_3)_3 \cdot 9 \text{H}_2\text{O}$) to above solution.
- C. Add hexone. Mix solutions together.
- D. Centrifuge.
- E. Transfer off hexone (upper) layer after centrifuging, with a transfer pipette, to a platinum dish.
- F. Evaporate solution to dryness.
- G. Fuse residue with sodium fluoride - lithium fluoride salt.

- H. Measure amount of fused residue fluorescence with fluorometer to obtain gross uranium fluorescence.
- I. Convert to amount of uranium from fluorescence calibration curve for known amounts of uranium.

The alpha detector used in this study is a gas-flow ionization counter. The block diagram of the detection system is illustrated in Figure 13.

An alpha particle passing through the chamber leaves a trail of dense ionization, consisting of electrons and positive ions. The electrons will migrate towards the negative electrode and the positive ions will migrate towards the positive electrode.

electric field of the chamber. The electrons are collected at the negative electrode and the positive ions are collected at the positive electrode.

The ionization current is measured by a sensitive electrometer. The ionization current is proportional to the number of alpha particles passing through the chamber.

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APPENDIX C

AN ALPHA COUNTER TO DETECT PLUTONIUM CONTAMINATION*

Theory of Operation

The alpha detector used in this study is a gas-flow ionization counter. The block diagram of the detection system is illustrated in Figure 13.

An alpha particle passing through the chamber leaves a track of dense ionization, consisting of electrons and positive ions. The electrons will migrate towards the anode, and the positive ions will migrate towards the grounded detector walls. As the ions move in the electric field of the electrodes, charges are induced on the electrodes. An alpha particle passing through argon gas loses about 26 eV per ion pair produced.⁽¹⁶⁾ Assuming that all of the alpha particle's energy is lost in ionizing the gas, about 2.0×10^5 ion pairs will be produced by a 5.1 MeV plutonium alpha particle. If the charge sensitivity of the preamplifier is $0.16 \mu\text{V}$ per ion pair** and if the amplifier gain is about 1000, the output pulse, produced by a plutonium alpha particle should be about 3 volts. This pulse could then be viewed with an oscilloscope or could be fed into a voltage discriminator and counter with a scaler.

Beta particles have a much lower specific ionization potential than do alphas. As Figure 14 illustrates, in the ionization

*designed by Glenn Brunson, EBR-II Project, Analysis and Physics Div.

**for a Tennelec FET preamplifier, TC 133.

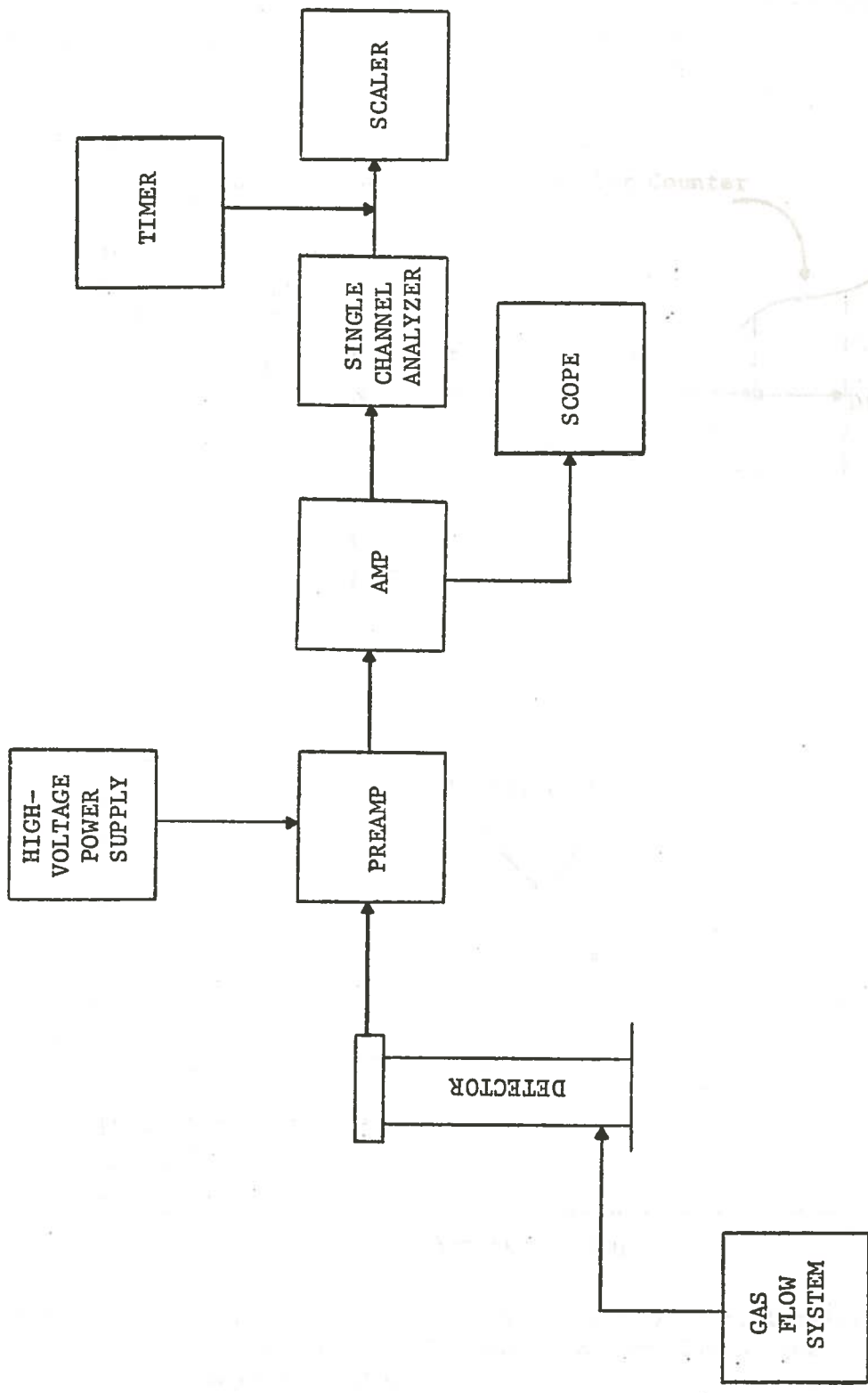


FIGURE 13. Block Diagram of Detection System.

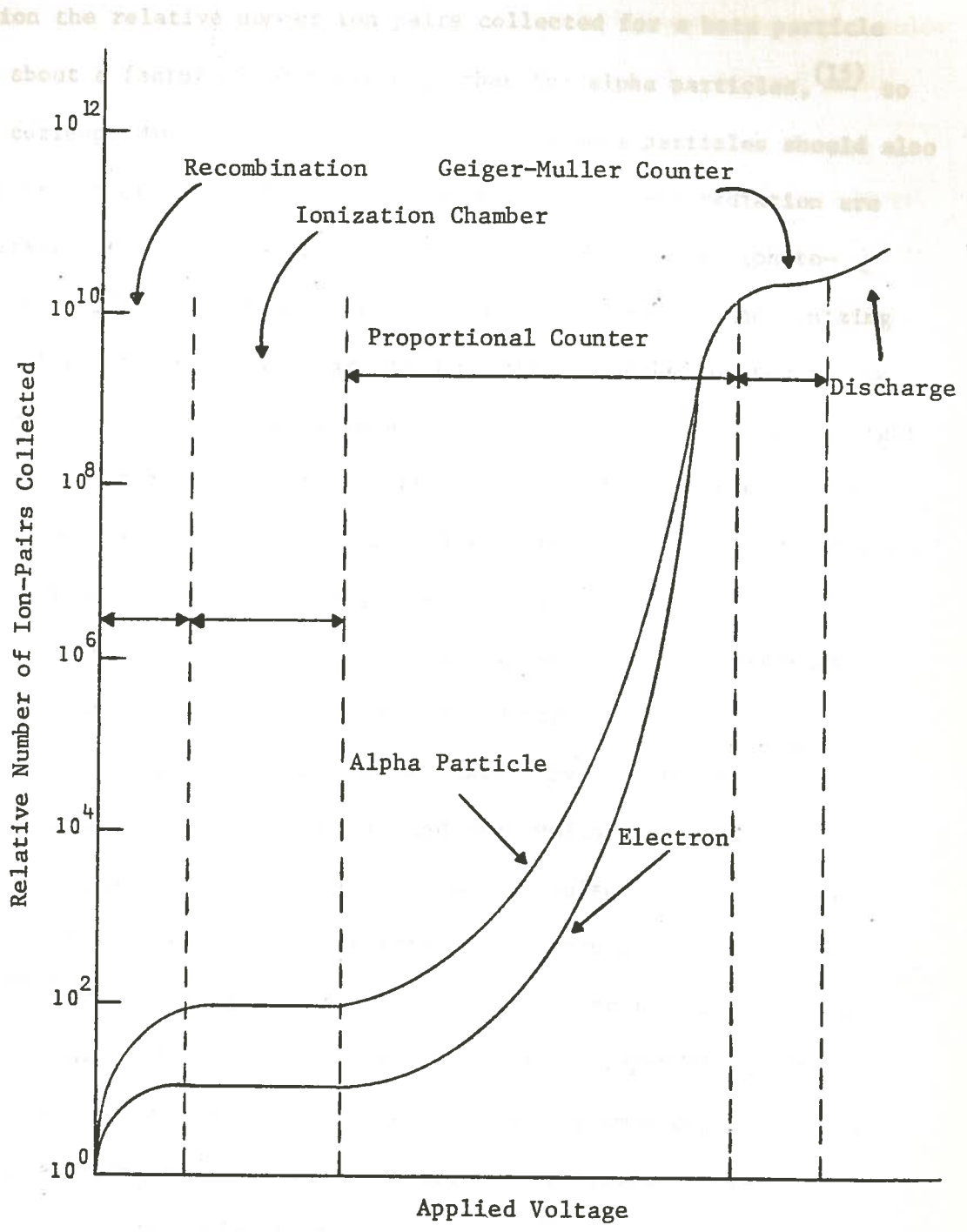


FIGURE 14. Illustration of the Relative Number of Ion-Pairs Collected in a Counter as a Function of the Applied Voltage.

region the relative number ion pairs collected for a beta particle are about a factor of 10 lower than that for alpha particles, ⁽¹⁵⁾ so the corresponding pulse heights produced by beta particles should also be a factor of 10 smaller. Pulses produced by gamma radiation are generally less troublesome because an intervening event (photoelectric effect, Compton effect) is required to produce an ionizing particle. This suggests that alpha particles can be counted in the presence of large fluxes of beta and gamma radiation. A pulse height discriminator can be set to reject low amplitude pulses and a scaler used to tally the alpha particle pulses. However, background levels of beta and gamma radiation can become so high that the super-position of their pulses results in signals of sufficient size and frequency to interfere with the counting of the alpha particles. ⁽¹⁶⁾

The operating point for an ionization pulse counter is dependent on the amplifier, discriminator, and high voltage settings. The amplifier settings determine both the output pulse size and shape. The high voltage settings also determine the output pulse size. The discriminator settings, on the other hand, determine which pulses will be counted. Thus, the steps in evaluating the operating point for the ionization counter are: (1) determining the optimum amplifier settings, (2) determining the optimum discriminator settings, and (3) determining the optimum high voltage setting.

The optimum amplifier settings are dependent on the voltage of the output pulses leaving the detector chamber. With the high voltage

fixed at 1000 volts, an oscilloscope is used to observe the alpha pulse heights. The amplifier gain is then varied until the desired pulse heights are obtained.

To determine the optimum discriminator settings the high voltage is again fixed, and the count rates are recorded for different discriminator settings. For too low a discriminator setting pulses produced by alpha particles and those produced by other sources will be counted (such as beta particles and electrical noise). For too high a setting, none of the alpha pulses will be counted. As Figure 15 illustrates, a range of discriminator settings is obtained, over which there is little variation in the observed count rate. In this region, most of the alpha pulses are counted while few of the extraneous pulses are counted. The ideal discriminator setting is on this plateau, near the high discriminator voltage end. This setting will provide maximum discrimination of noise and extraneous pulses.

The final task is to determine the optimum high voltage setting. With the discriminator setting fixed, the high voltage is varied and the corresponding count rates are recorded. At low voltages, many of the ionized particles that are produced do not reach the electrodes but recombine in the gas. As the voltage is increased, a sufficient number of electrons freed by the most energetic alpha particles reach the anode, producing pulses which pass the discriminator (recombination region). As the high voltage is further increased, a point is reached where all of the alpha particles produce pulses of sufficient height to

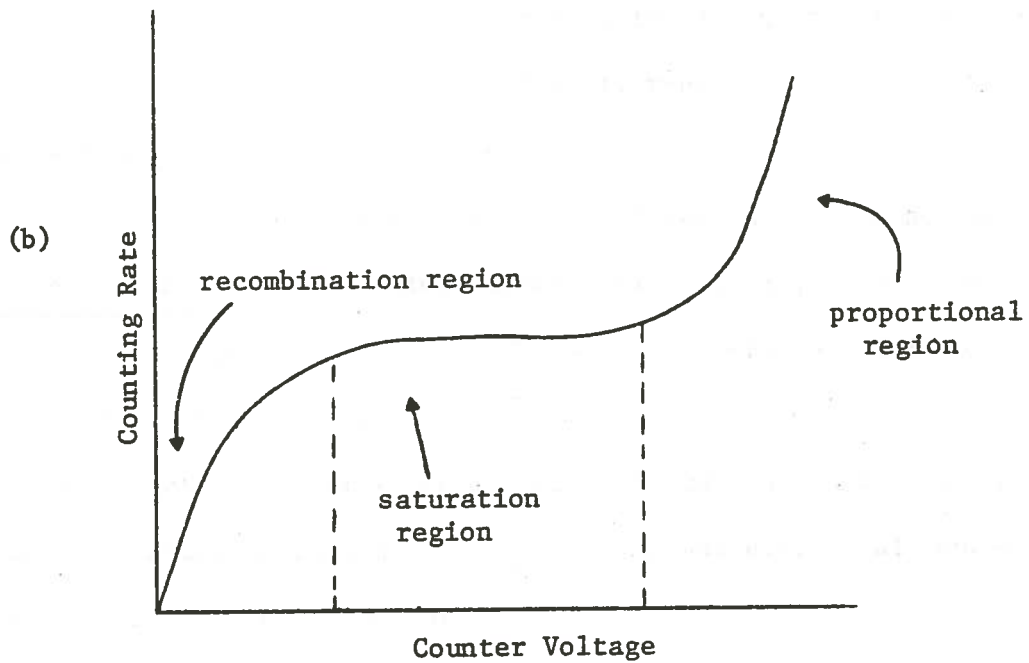
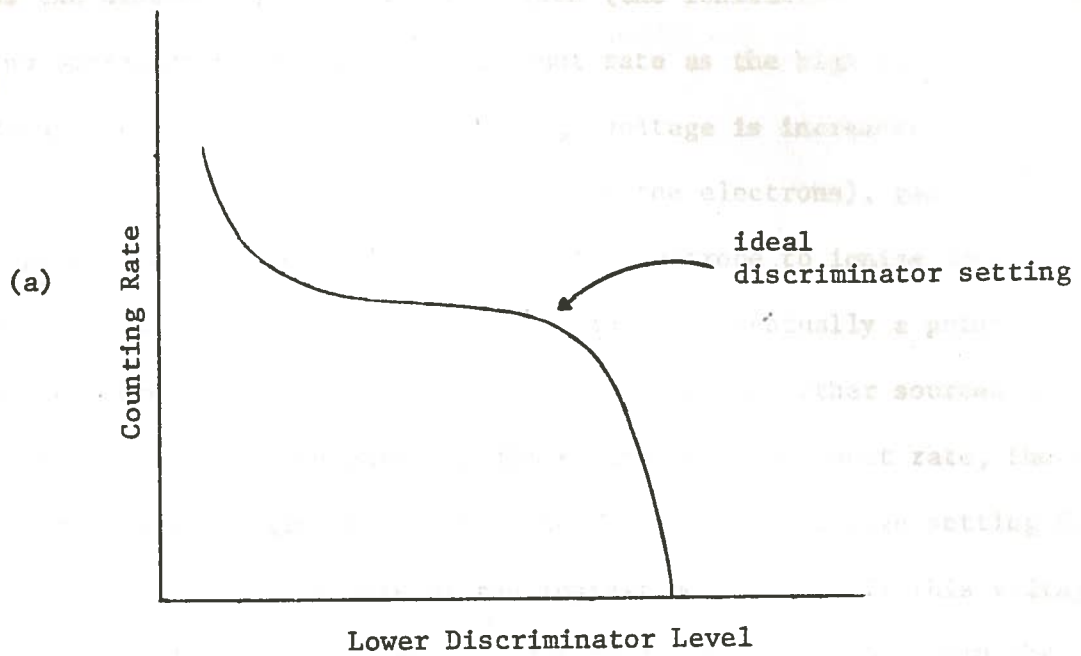


FIGURE 15a and b. Detector Response Curves. (a) Counting rate versus lower discriminator level. (b) Counting rate versus detector voltage.

pass the discriminator. In this region (the ionization region) there is no appreciable increase in the count rate as the high voltage is increased (see Figure 15). As the high voltage is increased beyond this region, the ions produced (primarily the electrons), gain enough energy as they move towards the positive electrode to ionize additional gas atoms in the interelectrode space. Eventually a point is reached where pulses produced by beta particles and other sources are of sufficient height to pass the discriminator. The count rate, therefore, once again begins to climb. The optimum high voltage setting for the chamber is in the middle of the ionization region. If this voltage is markedly different from the voltage originally used to obtain the discriminator setting, another discriminator vs. count rate plot is run for this voltage, and the whole procedure for evaluating the optimum discriminator and high voltage settings is repeated.

Construction of the Alpha Counters

Two alpha counters were built; one for testing in the hot cell environment and one for testing out-of-cell. The major parts of each counting system were the detector chamber, the detector electronics, and the detector gas system.

The detector chamber shown in Figure 16 consisted of three major parts: a cylindrical cathode, a detector cap and electrical connections, and a detector base plate.

The detector cathodes were constructed of aluminum tubing. Aluminum was selected over other materials on the basis of cost,

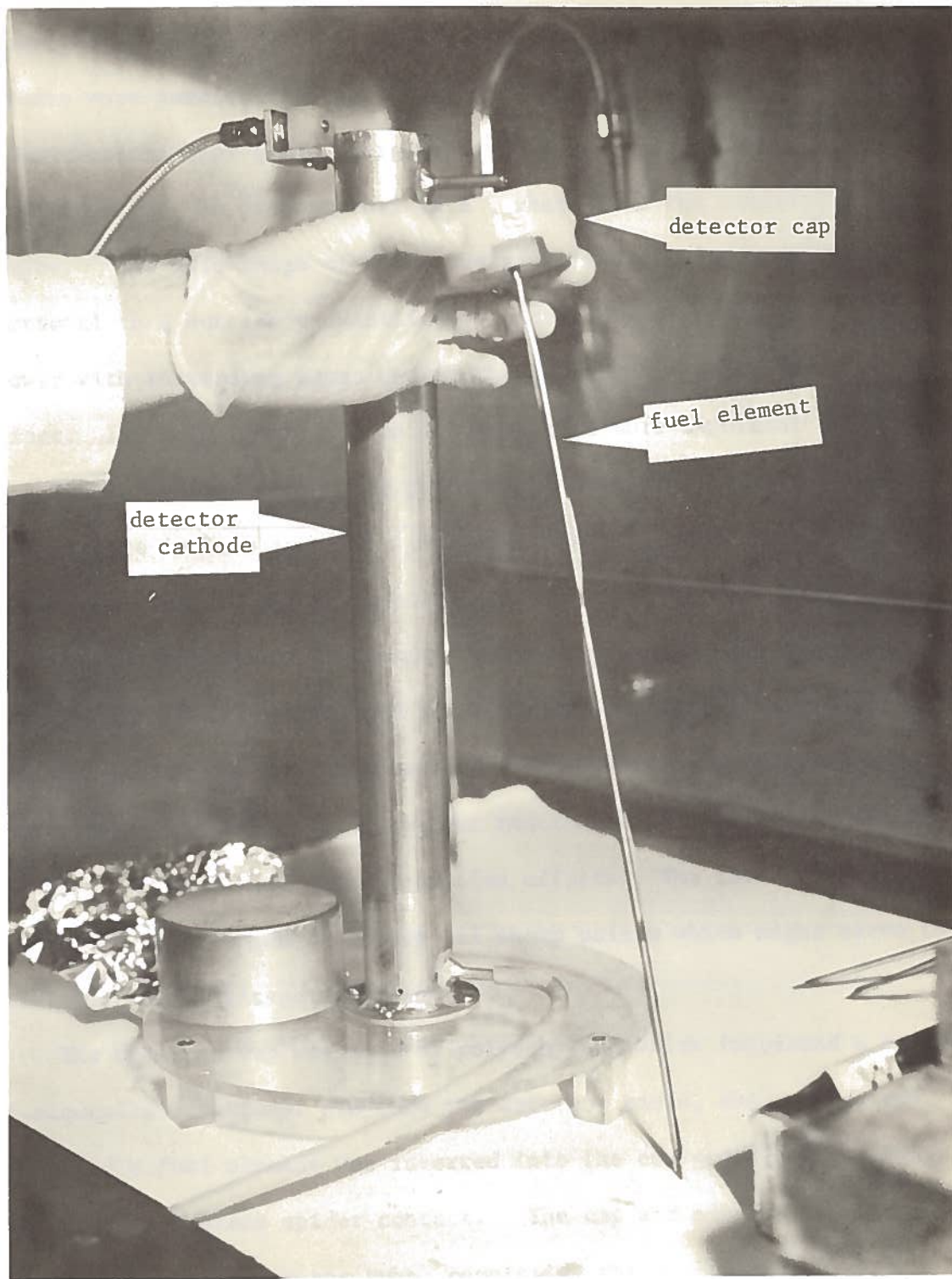


FIGURE 16. The Detector Chamber.

availability, fabrication, and the photoelectric, Compton, and pair production effects. Photoelectric, Compton, and pair production effects were particularly important to the operation of the in-cell alpha counter. For gamma energies between 0 and 3 MeV, the photoelectric effect is proportional to Z^5 , where Z is the material atomic number. For this range of gamma energies, the Compton effect is proportional to Z but its probability of occurrence decreases much more slowly with increasing gamma energy than does the photoelectric effect. In the range of 0.6 to 2.5 MeV it is more important than the photoelectric effect. ⁽¹⁷⁾ The probability of pair production in this energy range is less important than either of the other two effects. To minimize the probability of these processes occurring, a low Z material was selected (aluminum). The tubing for the out-of-cell detector was 18-1/2-inches high by 2-inches diameter. The in-cell detector tubing was 18-1/2 inches by 1-1/4 inches. The smaller diameter tubing was selected to further reduce the probability of photoelectric, Compton, and pair production effects. The inner wall of each tube was polished to remove all sharp points which might serve to initiate a high voltage discharge.

The detector cap was made of polyethylene which insulated a series of electrical contacts. To form the detector anode, the cap was removed and a fuel element was inserted into the cap guide hole, making contact with a brass spider contact. The cap and element were then inserted into the detector tube, completing the electrical circuit.

The anode was insulated from ground by a polyethylene centering cone. Each detector was fastened to a plexiglass base plate.

The basic electronics of both the in-cell and out-of-cell detection systems are listed in Table XII. These components were arranged as shown in Figure 13. Electrical cables in the detection system were originally all RG58A/U shielded cables (capacitance of 30 pf per foot). This cable was later changed to RG71/U shielded cable (capacitance of 13 pf per foot) to reduce the background counting rate.

The argon gas system consisted of a bottle of pure argon gas, a gas flow regulator with flow meter, and the gas transfer tubing. Out-of-cell, a 10-foot section of 1/4-inch diameter surgical rubber tubing transferred argon to the 1/4-inch diameter inlet tube of the detector. In-cell, a 1/8-inch stainless steel line provided the gas feed to the detector. The line was attached to a quick disconnect adapter on the gas inlet tube. The flow meter provided variable gas flow settings between 0 and 40 cubic feet per hour.

Out-of-Cell Testing and Results

Both detectors went through a similar series of out-of-cell tests. Tests included were: (1) the determination of operating point parameters, (2) the determination of counter efficiencies and background counting rates, and (3) special tests.

The detector operating point parameters were determined as was previously outlined. (See Theory of Operation section.) Pulse heights in the range of 3 to 10 volts were observed for an amplifier gain of

1000. Fluctuations of the discriminator as well as
rate of the counter were also that were obtained

TABLE XII

BASIC ELECTRONICS OF THE ALPHA COUNTER

-
- Tennelec, Model TC 940, D.C. High Voltage Power Supply
 - ORTEC, Model 401A, Instrument Bin and Power Supply
 - Tennelec, Model TC 550, Scaler
 - ORTEC, Model 406A, Single Channel Analyzer
 - Tennelec, Model TC 203BLR, Linear Amplifier
 - Tennelec, Model TC 133, FET Preamplifier
 - Cramer Controls Corporation, Type 117, Timer
-

1000. Figure 17 illustrates the respective discriminator vs. count rate and high voltage vs. count rate plots that were obtained for each detector. A discriminator setting of 2.0 volts was selected for the 2-inch detector, while a discriminator setting of 1.5 volts was selected for the 1-1/4-inch detector. Operating point voltages of 300 and 100 volts were chosen for the 2-inch and 1-1/4-inch detectors respectively. Ideally, as Figure 18 illustrates, high voltages closer to 500 volts should have been selected. However, sporadic discharging was observed for voltages greater than 800 volts. The lower voltages were selected because of this arcing problem, and because the count rate plateaus showed negligible slopes. A 50,000-dpm U-233 source was used to determine the operating point parameters. The U-233 activity (alpha energies about 4.8 MeV) was deposited onto the surface of a fuel element, and fixed to the element surface by a very light coating of lacquer.

The detector efficiencies were determined for two alpha standards: a 50,000-dpm U-233 tagged element and a 2000-dpm element. Knowing the alpha activity of each standard, the detector efficiency was determined from the equation:

$$\text{Efficiency} = \frac{\text{observed standard alpha activity}}{\text{actual standard alpha activity}}$$

For the 2-inch detector, efficiencies of 0.45 and 0.49 were observed for the 50,000 and 2000 dpm standards, respectively. Identical efficiencies were observed for the 1-1/4-inch detector. The detector

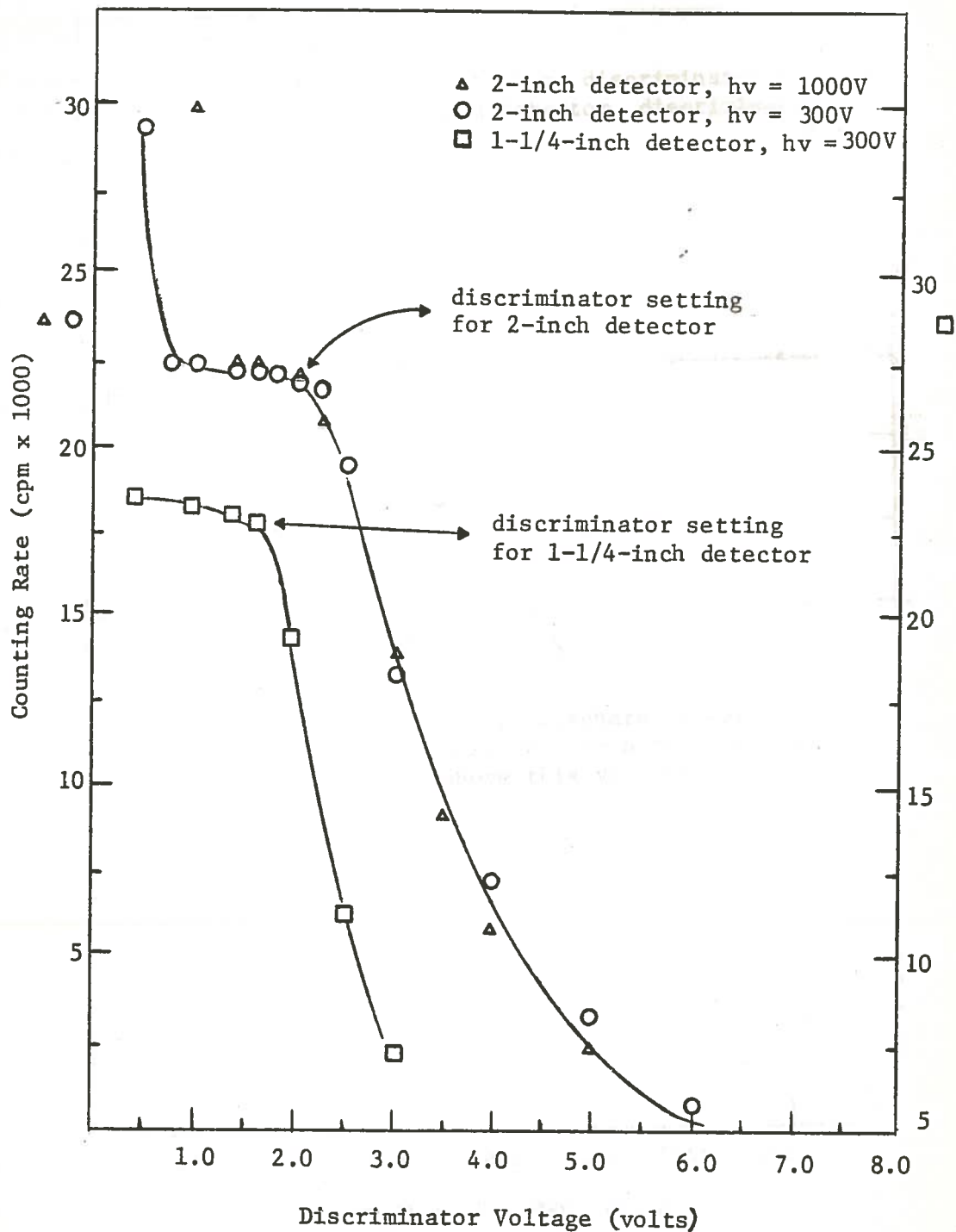


FIGURE 17. Counting Rate Versus Lower Discriminator Level for 2-inch and 1-1/4-inch Detectors.

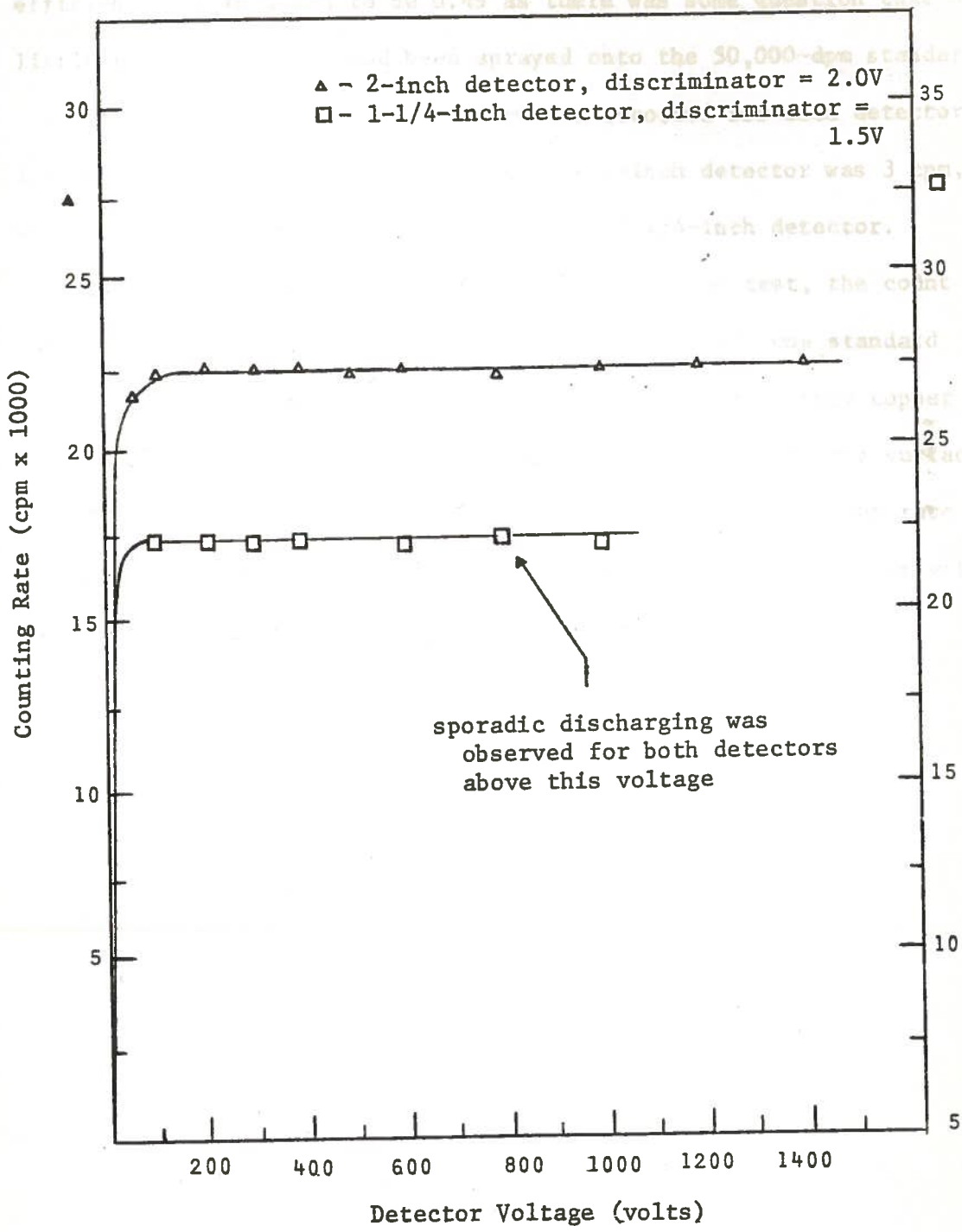


FIGURE 18. Counting Rate Versus Detector Voltage for 2-inch and 1-1/4-inch Detectors.

efficiencies were taken to be 0.49 as there was some question that a little too much lacquer had been sprayed onto the 50,000-dpm standard.

Five 5-minute background counts were recorded for each detector. The average background count rate for the 2-inch detector was 3 cpm, while a rate of 6 cpm was recorded for the 1-1/4-inch detector.

Two special tests were also performed. In one test, the count rates for two 2000-dpm alpha standards were compared, one standard with a spacer wire and one without. In another test, a tiny copper wire, coated with Co-60 (about 100 mR/hr) was fastened to the surface of a 2000-dpm standard, and its effect on the standard's count rate was noted. Neither the spacer wire nor the added beta-gamma activity had any effect on the observed standard count rates.

VITA

Bruce Michael Durst was born on June 29, 1949, in New Orleans, Louisiana, the third of four sons born to Mary Rita and Robert Stevenson Durst. He attended Edmonds Senior High School in Edmonds, Washington, and graduated in 1966.

In 1966, he entered the University of Washington, in Seattle, and graduated with a B.S. in physics in June of 1970. After teaching at Edmonds Community College for half a year, he entered graduate school in the field of astronomy at Louisiana State University, receiving a teaching assistantship in the same field.

He decided in May of 1972 to pursue graduate studies in nuclear engineering at LSU, and received a graduate assistantship in this field.

He was awarded an Argonne National Laboratory fellowship in May of 1973 to pursue his graduate research at the Experimental Breeder Reactor-II in Southeastern Idaho.

Mr. Durst was married to Kathryn Lee Fish on July 16, 1971. They have one daughter, Heather (2).