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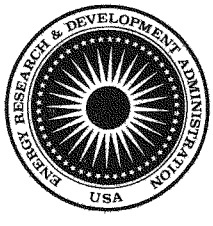
Dale R. Schaeffer, Ernest W. Johnson, Warren E. Sheehan, Donald L. Fleming and Eugene E. Egleston

November 22, 1976



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Abstract

$^{238}PuO_2$ contamination of the grit blasted surface of the primary impact shell assembly (PISA) of the multi-hundred watt isotopic heat source was measured. The study determined the amount and distribution of the $^{238}PuO_2$ and characterization of its behavior during aging at 1350°C. The results concluded that normal decontamination effectively removes the superficial $^{238}PuO_2$ but does not extract the $^{238}PuO_2$ which is deep within the grit blasted structure. Subsequent heating results in migration of microcurie amounts of plutonium out of the grit blasted structure.

Introduction

The containment of the plutonium used in the multi-hundred watt isotopic heat source is of utmost importance in providing an environmentally safe product. Stringent limits have been placed not only upon the containment of the plutonium within the primary iridium container (called the Post Impact Shell Assembly or PISA), which is shown in Figure 1, but also on the amount of plutonium remaining on the outside surface of the PISA after it is fabricated. Decontamination of the iridium surface to well below the acceptable limit is readily accomplished at Mound Laboratory but a test was designed to assess the behavior of the small amount of the remnant surface plutonium as a function of time at operating temperature. The test involved heating the PISA for various periods of time at 1350°C (the service temperature) and characterizing the amount of distribution of the plutonium on the surface after each heat treatment. After a total of 1535 hours at temperature, the distribution of plutonium in the cross-section of the iridium wall of the PISA was determined by removing layers of iridium from the surface by electropolishing and determining the plutonium content in each layer.

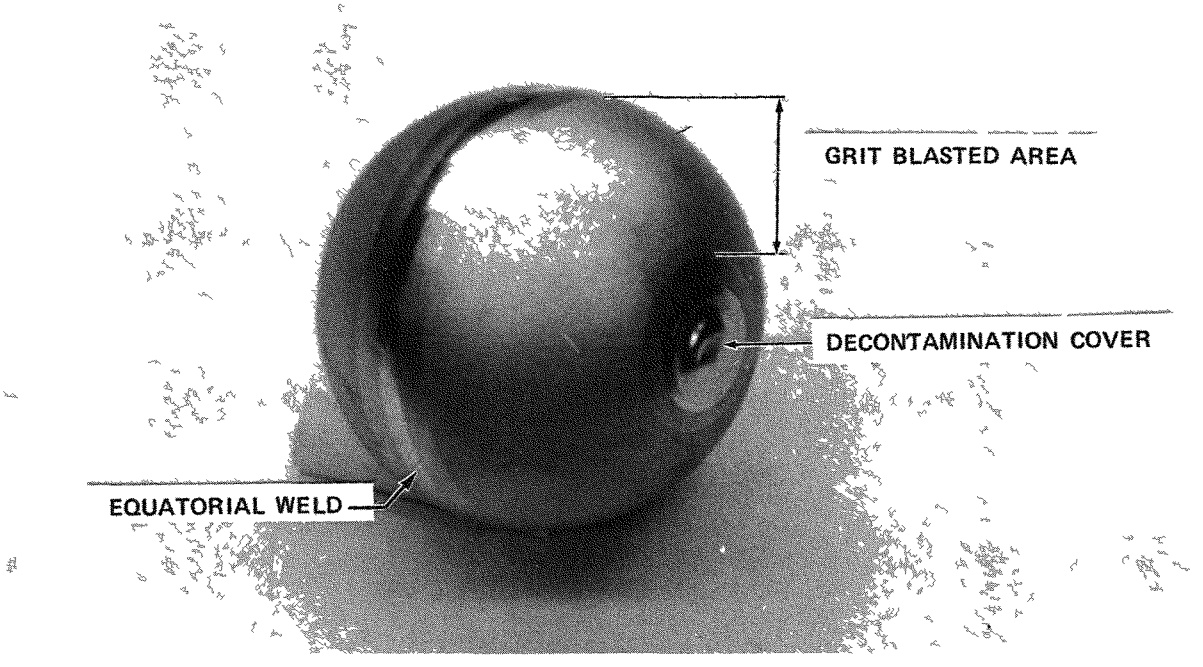

FIGURE 1 - Photograph of the primary impact shell assembly (PISA). It consists of two iridium hemishells which, when welded together, form the container for a $^{238}PuO_2$ sphere.

Experimental

The PISA used in this test is shown in Figure 1 and was fabricated in the "hot" facilities normally used in production. It was fabricated in the same manner as the production PISA's with the exception that just prior to welding, the $^{238}PuO_2$ fuel sphere was replaced by a ThO_2 simulant sphere. After fabrication, the $^{238}PuO_2$ was removed (decontaminated) from the outside surface of the PISA by the normal procedure of soaking in boiling acid (50% HF/50% HNO_3), rinsing with water, scrubbing with scouring powder and water, and rinsing again with water. This decontamination procedure was repeated three times. The PISA was then helium leak checked and examined for surface alpha contamination. Measurements of the surface alpha contamination were made by first wiping the surface with a piece of cloth (called a swipe) and measuring and, thus, obtaining an indication of the amount of loose or wipeable $^{238}PuO_2$ present on the surface. The second method of determining the alpha contamination involved direct measurement of the alpha radiation emitted from various portions of the PISA. The PISA was then inserted into a graphite impact shell (GIS), which is shown in Figure 2. The decontamination covers (see Figure 1), which are normally removed prior to installation in the GIS, were left intact to eliminate the possibility of $^{238}PuO_2$ coming out of the vents, which are underneath the decontamination covers, and to allow future decontamination, leak checking, and dye penetrant examination. The PISA and surrounding GIS were then subjected to heat treatments at 1350°C in a vacuum furnace for various periods of time up to a total of 1535 hours.

After each heat treatment, swipe measurements of the alpha contamination were made of the PISA, GIS, and furnace. Direct determinations of the alpha contamination were also made on the PISA and GIS after each heat treatment but unlike the swipe measurements, they were made after the PISA and GIS were again decontaminated. Decontamination of the PISA involved scrubbing with scouring powder and water* with the resultant mixture examined for its plutonium content by

*The scouring powder and water decontamination of the PISA differs from the initial acid decontamination in that the scouring powder and water removes only the surface plutonium, whereas the boiling acid decontamination dissolves the $^{238}PuO_2$ and is thus able to remove it from deeper within the grit blasted structure.

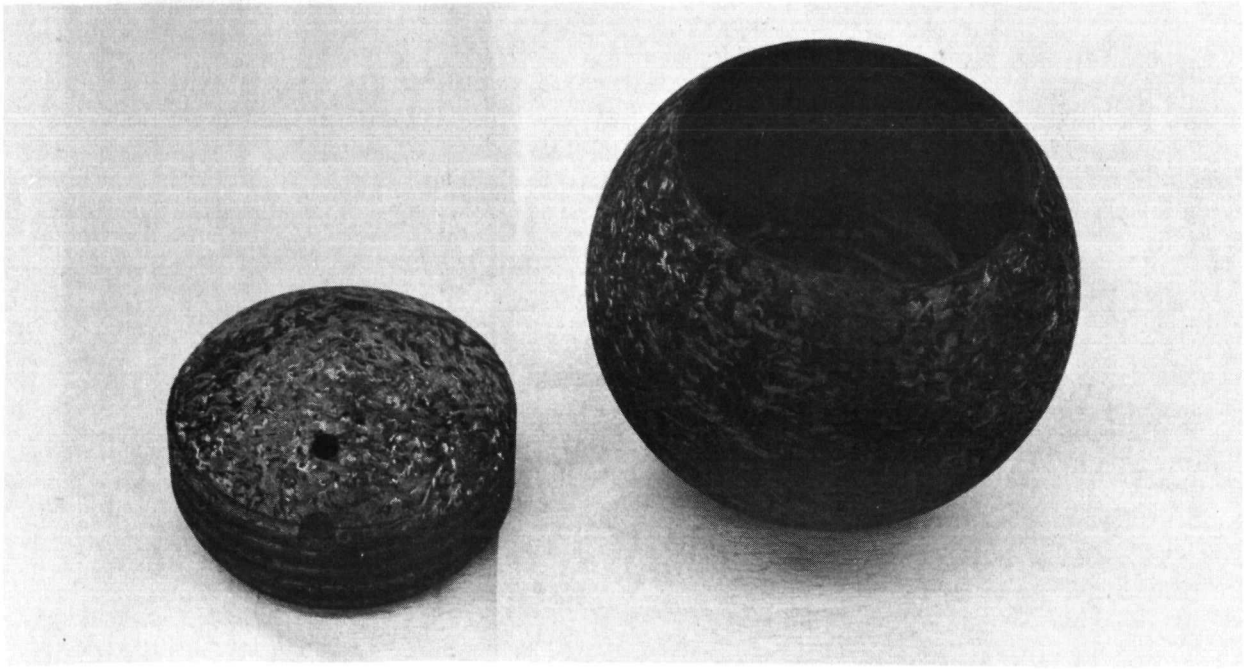

FIGURE 2 - Photograph of the graphite impact shell (GIS) which is used to contain the PISA. After the PISA is placed into the cavity of the body of the GIS, the cap is screwed into the body thereby providing impact protection.

liquid scintillation. The GIS was decontaminated by wiping it with a dry rag. After each heat treatment, the PISA was also helium leak checked and after all but the first heat treatment (100 hr at 1350°C) the PISA was examined for surface defects by dye penetrant.

After a total of 1535 hr at temperature, the distribution of plutonium in the cross-section of the iridium wall in the grit blasted area was measured. This was accomplished by removing "layers" of iridium from the outside surface of the PISA by electropolishing and determining the plutonium content in the removed layer by finding the amount of plutonium in the electrolyte used to remove the layer. The plutonium distribution was also characterized by measuring the direct alpha radiation emitted from the newly exposed surface.

Removal of iridium from the surface of the PISA was accomplished by clamping the PISA into the fixture described in Figures 3 and 4, which allowed a portion (0.11 $in.^2$) of the PISA's outer surface to be in contact with an electrolyte held in a reservoir at the top of the fixture. Ten volts were then applied between the PISA and an electrode submerged in an electrolyte consisting of 130 gm of KCN in one liter of water. A depth micrometer was used to measure the thickness of the removed iridium layer by measuring the distance between the top of the fixture and the "top" of the PISA. The electropolishing procedure resulted in the removal of about 0.1 mil after 5 min of polishing. A photograph of the PISA after removal of a portion of its surface is given in Figure 5.

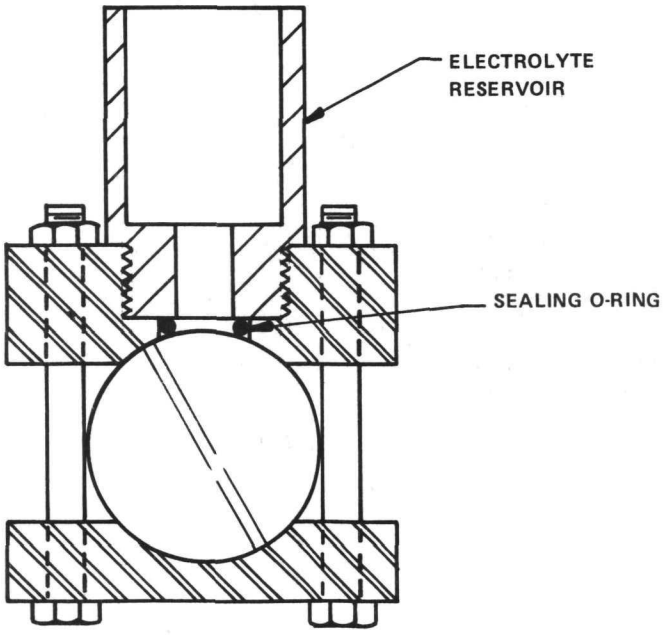

FIGURE 3 - Sketch of test fixture used to remove a portion of the PISA surface.

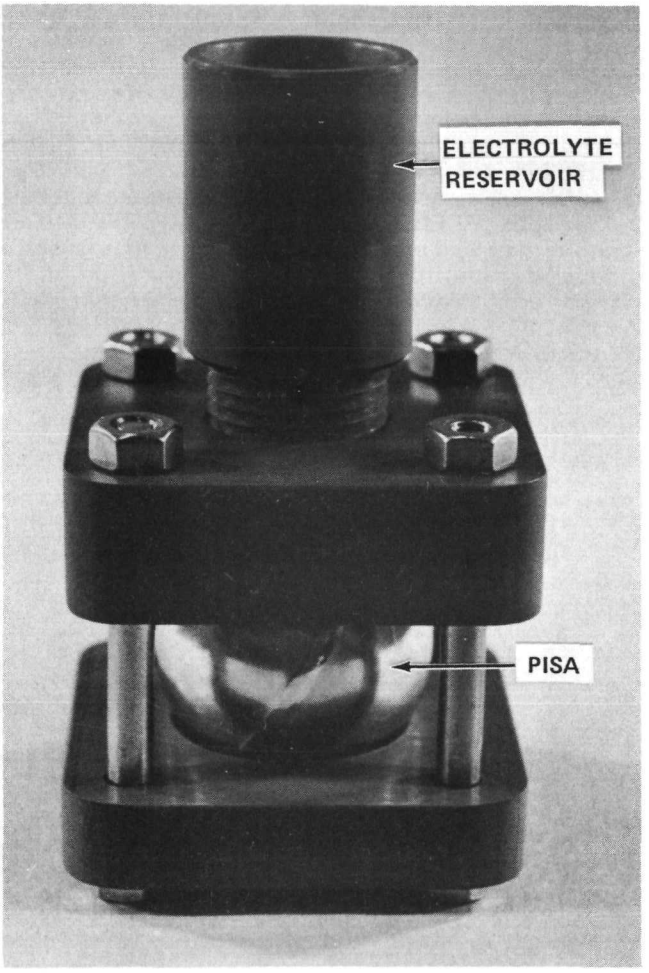

FIGURE 4 - Photograph of the test fixture used to remove portions of the PISA surface.

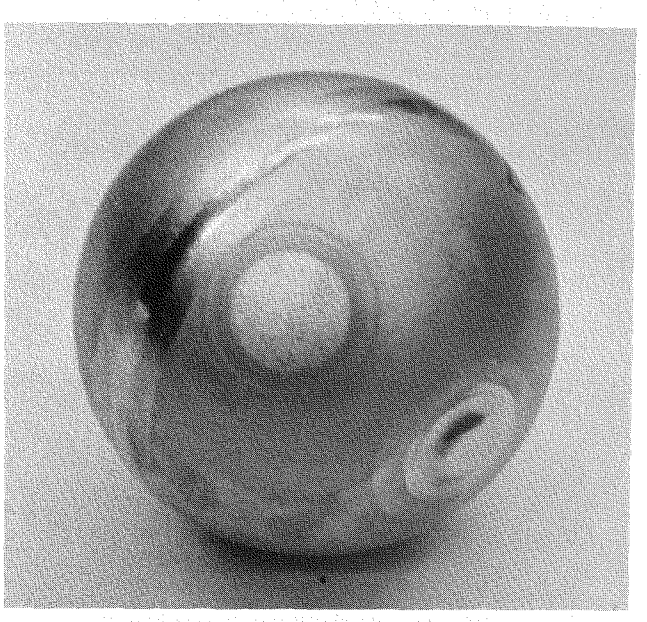

FIGURE 5 - Photograph of the PISA after removal of 0.0007 in. from a portion of the surface. Note that the grit blasted structure is gone and that the grain structure is now visible.

After electropolishing the iridium surface, the electrolyte and subsequent reservoir washings were submitted for liquid scintillation assay to determine their plutonium content and, thus, the plutonium content of the removed surface layer. Measurements of the direct alpha radiation emitted from the newly exposed iridium surface were accomplished by removing the reservoir and sealing O-ring and replacing them with a new O-ring and mask. This setup is shown in Figure 6. The O-ring and mask insure that only the newly exposed surface was measured for alpha radiation. The direct radiation measurements of the iridium surface were relative rather than quantitative measurements of the amount of plutonium present on the surface.

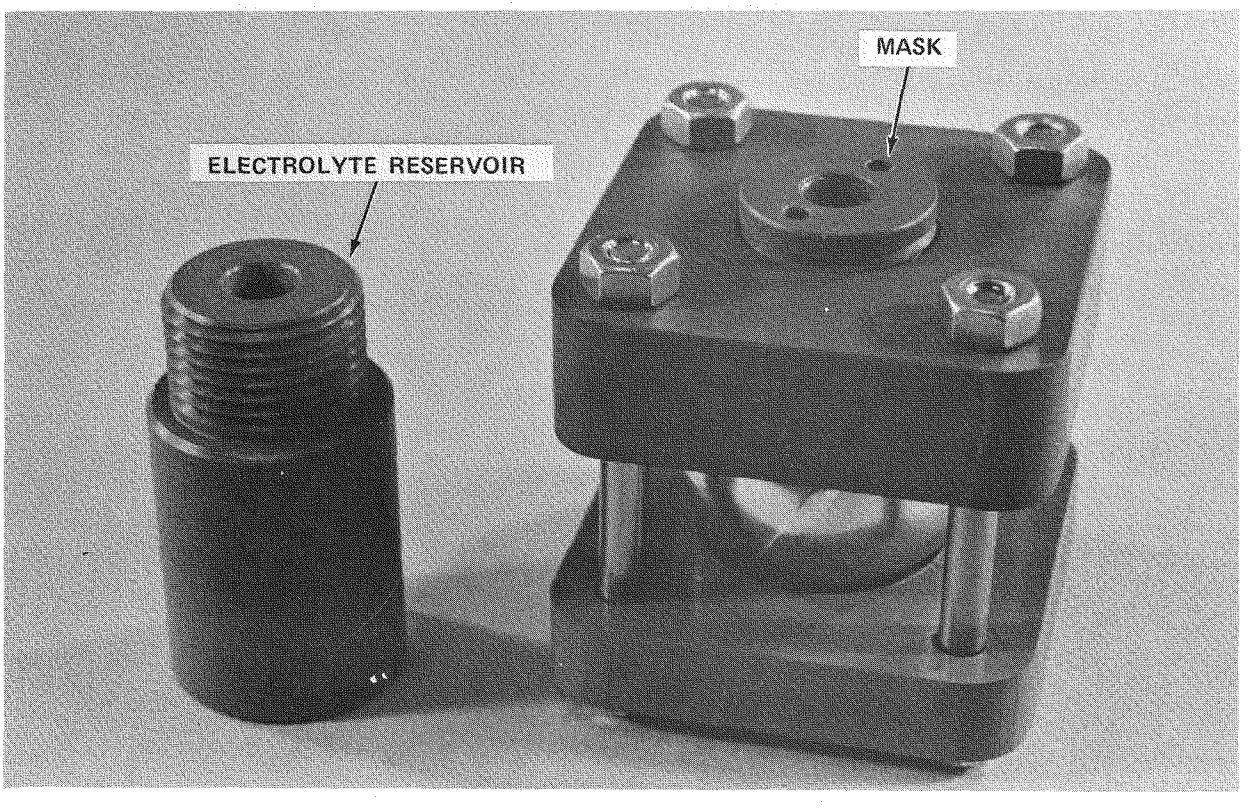

FIGURE 6 - Photograph of the test fixture used to measure the direct alpha radiation emitted from the surface of the PISA after a portion of the surface had been removed.

Results

A substantial increase in the alpha contamination on the external PISA surface occurs with time at temperature as proven by increases in both wipeable and direct alpha radiation. The data obtained from the swipes of the PISA are compiled in Table 1 and the averages of the data are plotted versus time at temperature in Figure 7. The largest increase in wipeable alpha contamination occurred in the two grit blasted areas (see designated regions in Figure 1) located between the decontamination covers and the equatorial weld. Table 2 contains the results from the measurements of the direct alpha radiation emitted from the various portions of the PISA and the averages of these data are displayed in Figure 8. The greatest increase of direct alpha radiation was from the general area of the decontamination covers with the second largest increase from the grit blasted areas. Further direct measurements of both decontamination cover areas using masks to isolate the radiation emitted from the various regions in the area found the decontamination covers, the non-grit blasted areas encircling the decontamination covers, and the surrounding grit blasted areas to emit 3%, 15%, and 82% of the direct alpha radiation, respectively. Therefore, the grit blasted areas of the PISA experienced the largest increase in direct as well as the wipeable alpha radiation.

Liquid scintillation measurements of the alpha radiation emitted from the water/scouring powder mixtures used to decontaminate the PISA were 2×10^6, 4.03×10^7, and 4.96×10^7 DPM after the 300,700, and 1535 hr treatments, respectively. The total of 9.2×10^7 DPM corresponds to 41.4 μCi of plutonium which was removed from the surface. An additional 0.9 μCi was removed by the swipes making the total amount of plutonium which was removed roughly 42.3 μCi.* A graph of the amount of plutonium which was removed versus time at temperature is given in Figure 9.

The determination of the cross-sectional distribution of plutonium in the PISA wall in the grit blasted area found a concentration of plutonium at the exterior edge. Table 4 lists the direct alpha radiation measurements of the iridium surface at various depths into the wall. This information is displayed in Figure 10 and shows that the plutonium is concentrated in the grit blasted structure located on the outside 0.1 to 0.2 mils of the PISA wall. Table 5 lists the alpha radiation measurements of the electrolyte solutions used to remove the iridium surface layers. Figure 11 displays this data and reveals a similar plutonium concentration in the grit blasted structure as found during the direct alpha radiation measurements at various depths into the wall. The amount of plutonium contained in the grit blasted structure (the outer 0.15 mils), which was delineated by the sealing O-ring was about 0.92 μCi, which corresponds to approximately 55 μCi in the grit blasted structure of the entire PISA. Since about 42 μCi of ^{238}Pu were removed from the grit blasted structure during the 1350°C heat treatments and subsequent decontaminations, a total of roughly 97 μCi ^{238}Pu was originally in the grit blasted structure.

The plutonium which is indicated in Figures 10 and 11 as being in the iridium below the grit blasted structure is probably an anomaly, due to slow removal of the grit blasted structure underneath the O-ring used to seal the electrolyte reservoir to the PISA. However, the much less probable inward diffusion of plutonium from the grit blasted structure during the 1350°C heat treatments would cause a similar effect.

Dye penetrant examinations and helium leak checks of the PISA determined that the iridium sphere did develop faults in its exterior surface, but that the faults did not extend through the wall. The dye penetrant examinations revealed numerous pores on one end of the PISA in the area between the decontamination cover and the grit blasted area during the first examination which occurred after 300 hr at 1350°C, and in the same area on the other end of the sphere after 1535 hr. These pores were profuse, had a diameter of less than 0.001 in., and were deep enough to continue to strongly bleed dye. The dye penetrant examination did not find any other defects except after 1535 hr at temperature when several cracks were found in both of the decontamination covers and three 0.015-in. long crack indications were found about 0.050 in. from the equatorial weld. Helium leak checks of the PISA after all the heat

*42.3 μCi represents only the minimum amount of plutonium that was removed, since it was impossible to measure the plutonium remaining on the rags used for decontamination.

Table 1

WIPEABLE ALPHA CONTAMINATION OF PISA AS MEASURED BY SWIPES

Area	After Initial Decontamination and Before Heat Treatment (DPM)	After 100 Hr at 1350°C (DPM)	After 300 Hr at 1350°C (DPM)	After 700 Hr at 1350°C (DPM)	After 1535 Hr at 1350°C (DPM)
Decontamination Cover #1	14	285	2164	20,000	36,000
Grit Blasted Area Between Decontamination Cover #1 and Equatorial Weld	14	15	2096	12,000 to 20,000 16,000 Avg.	144,000 to 320,000 232,000 Avg.
Equatorial Weld	--	12 to 362 137 Avg.	1896 to 2986 2441 Avg.	12,000 to 16,000 14,000 Avg.	130,000 to 300,000 215,000 Avg.
Grit Blasted Area Between Decontamination Cover #2 and Equatorial Weld	20	25	2646	24,000 to 32,000 28,000 Avg.	300,000 to 400,000 350,000 Avg.
Decontamination Cover #2	20	350	2082	24,000	140,000

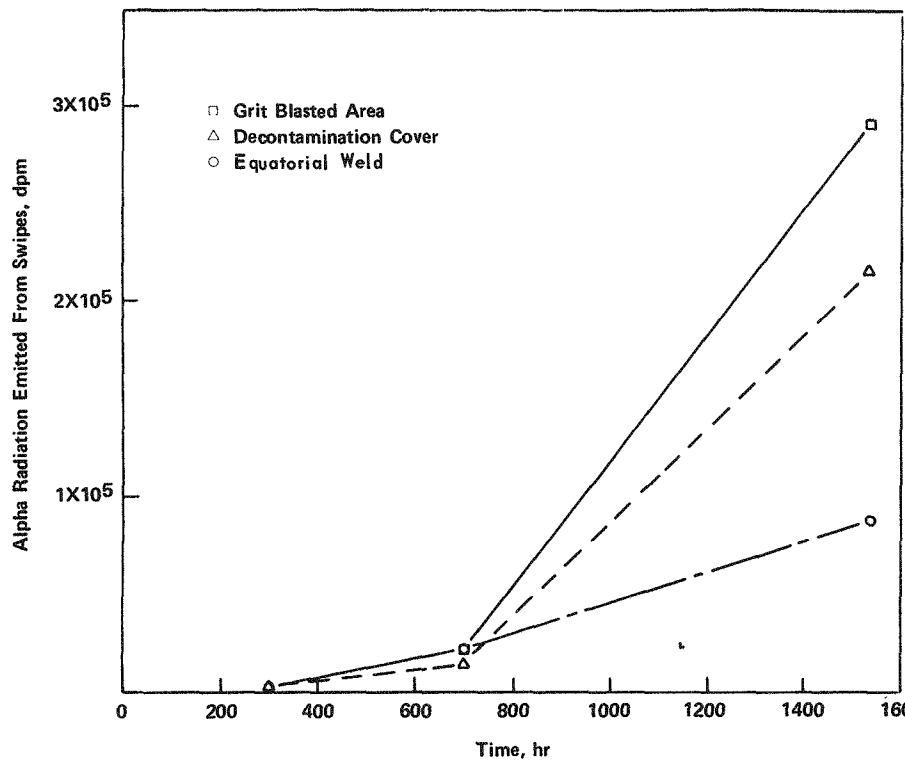

FIGURE 7 - Graph of the wipeable alpha contamination present on various regions of the PISA versus the time the PISA was exposed to 1350°C. The amount of wipeable alpha contamination was determined by taking swipes.

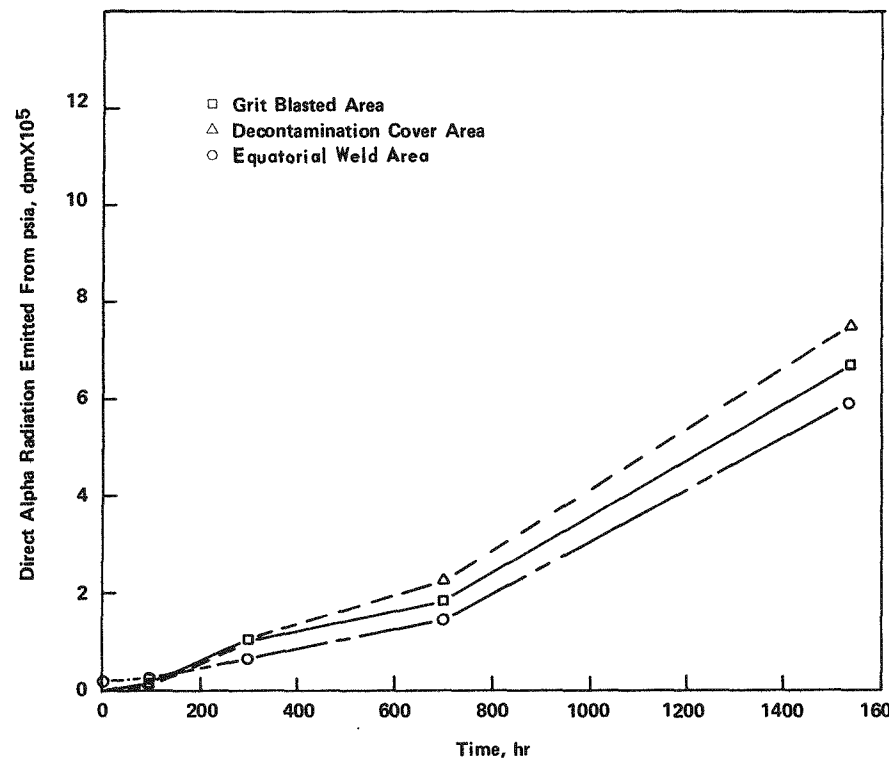

FIGURE 8 - Graph of the direct alpha radiation emitted from the various areas of the PISA versus the time the PISA was exposed to 1350°C.

Table 2

DIRECT ALPHA CONTAMINATION OF PISA[a]

Area	After Initial Decontamination and Before Heat Treatment (CPM)	After 100 Hours at 1350°C (CPM)	After 300 Hours at 1350°C (CPM)	After 700 Hours at 1350°C (CPM)	After 1535 Hours at 1350°C (CPM)
Decontamination Cover #1[b]	2	13,600	72,700	206,000	706,000
Grit Blasted Area Between Decontamination Cover #1 and Equatorial Weld	1,500	17,870	134,000	137,000	623,000
Equatorial Weld	4,700 to 47,000 18,300 Avg.	2,350 to 51,800 23,815 Avg.	18,700 to 119,800 64,600 Avg.	97,300 to 202,000 147,000 Avg.	480,000 to 732,000 598,000 Avg.
Grit Blasted Area Between Decontamination Cover #2 and Equatorial Weld	500	8,250	81,300	235,000	720,000
Decontamination Cover #2[b]	20	9,148	137,000	261,000	800,000

[a]Direct measurements of the alpha contamination were made after removal of the loose or wipeable plutonium with scouring powder and water.

[b]82% of the direct alpha radiation coming from the areas of the decontamination covers was emitted from the surrounding grit blasted region, while only 3% was from the decontamination cover. Therefore, the largest source of direct alpha radiation is the grit blasted regions, rather than the decontamination covers as indicated by this table.

Table 3

WIPEABLE ALPHA CONTAMINATION OF GIS AS MEASURED BY SWIPES

Area	Before Heat Treatment (DPM)	After 100 Hr at 1350°C (DPM)	After 300 Hr at 1350°C (DPM)	After 700 Hr at 1350°C (DPM)	After 1535 at 1350°C (DPM)
Internal Body	0	114	70	966	30,000
Internal Cap	0	4	48	168	10,000
Body Threads	0	8	10	80	6,000
Cap Threads	0	0	30	4	3,800
External Body*	0	30	20	4	3,800
External Cap*	0	12	22	6	0

*Alpha contamination on the exterior portion of the GIS after the shorter thermal exposures is probably due to the surrounding furnace which was wiping about 500 DPM prior to the test.

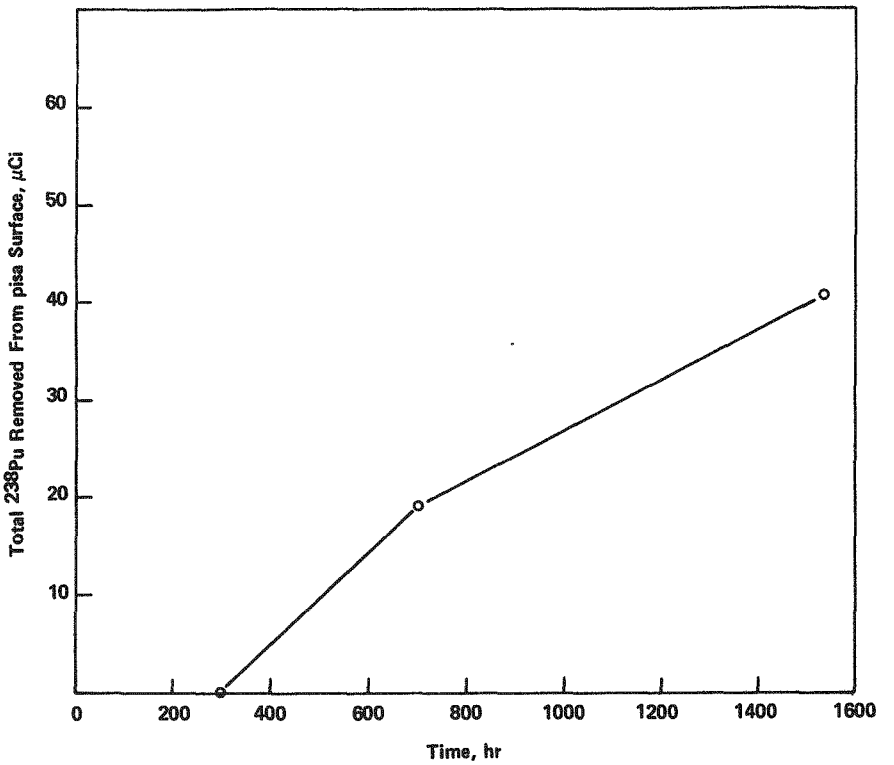

FIGURE 9 - Graph of the total amount of ^{238}Pu removed from the surface of the PISA as a function of the time the PISA was exposed to 1350°C. The ^{238}Pu was removed from the surface during decontamination and swipe measurements.

Table 4

DIRECT ALPHA RADIATION MEASUREMENTS OF PISA AT VARIOUS DEPTHS INTO THE WALL

Approximate Distance from Outer Edge of PISA (mil)	Direct Alpha Radiation Emitted from Surface (CPM)	Comments
0	6991	Electrolyte only in contact with PISA, no voltage applied.
0.1	1308	Grit blasted structure still visible after test.
0.2	521	Grit blasted structure gone, grain structure now visible.
0.3	275	
0.5	216	
0.7	185	

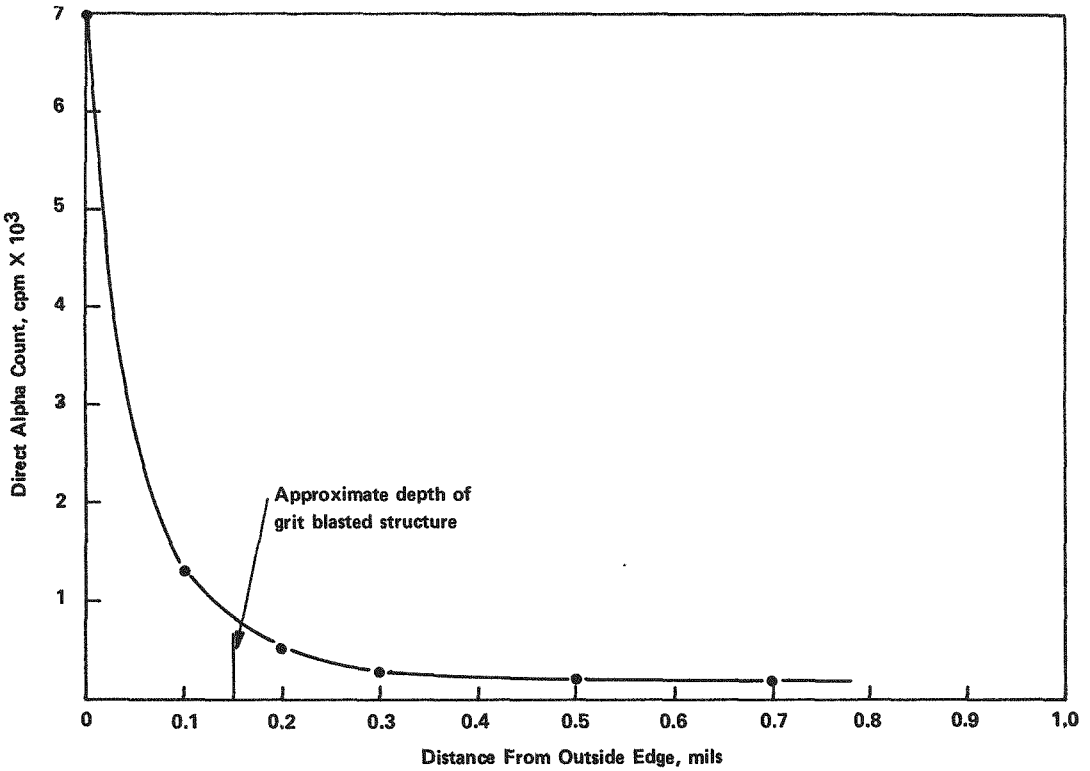

FIGURE 10 - Graph of the direct alpha radiation coming from the PISA surface as a function of depth into the wall.

treatments indicated that the capsule had retained its integrity, therefore, the defects found by dye penetrant did not extend through the capsule wall and cause the observed alpha contamination of the PISA surface.

Alpha contamination of the GIS was confined to the interior cavity which was in contact with the PISA and to the threads; there was no direct (the minimum limit of detection was 200 DPM) or wipeable alpha contamination detected on the outside of the GIS or in the vacuum furnace. Table 3 lists the wipeable contamination of the inside of the GIS. Direct alpha radiation emitted from interior of the GIS was detected only after the total of 1535 hr at 1350°C were completed. The direct readings prior to decontamination were 30,000 to 160,000 DPM from the cavity and 4000 DPM from the threads; and after decontamination (wiping with dry rag) were 6000 to 80,000 DPM from the cavity and 400 DPM from the threads.

Discussion

The level of wipeable and direct alpha radiation on the PISA surface just after fabrication and initial decontamination indicates that the amount of plutonium on and embedded in the surface is very low, however, subsequent aging of the PISA at the service temperature (1350°C) results in the increase of both wipeable and direct alpha contamination on the exterior surface. The source of this plutonium as determined by the area with the highest wipeable and direct alpha radiation was the grit blasted area. Proof that the grit blasted area was the source of the plutonium was obtained by determining the plutonium cross-sectional distribution in the PISA wall. This distribution was characterized by a concentration of plutonium in the grit blasted structure with little or no plutonium below it. The amount of plutonium in the grit blasted structure of the entire PISA was found to be approximately 55 μCi, which is slightly larger than the 42 μCi of plutonium which had been removed from the PISA surface during all of the 1350°C heat treatments; therefore, the grit blasted structure does contain enough plutonium to cause the observed surface contamination.

Table 5

ALPHA RADIATION MEASUREMENTS OF ELECTROLYTE USED TO REMOVE LAYERS OF PISA WALL

Distance of Layer from Outer Edge of PISA (mil)	Alpha Radiation Emitted from Electrolyte (DPM)	Pu Content of Marked Area (μCi)	Pu Content for PISA (μCi)	Comments
0	1.288×10^4	0.0058	0.35	Electrolyte only in contact with PISA, no voltage applied.
0 - 0.1	1.923×10^6	0.866	51.8	Grit blasted structure still visible after test.
0.1 - 0.2	2.523×10^5	0.114	6.8	Grit blasted structure gone, grain structure now visible.
0.2 - 0.3	1.010×10^5	0.0455	2.7	
0.3 - 0.5*	1.60×10^5	0.0721	4.3	
0.5 - 0.7*	1.175×10^5	0.0529	3.2	

*0.2 mil of the PISA wall was removed; therefore, the plutonium content per 0.1 mil is half of the amount given in the table.

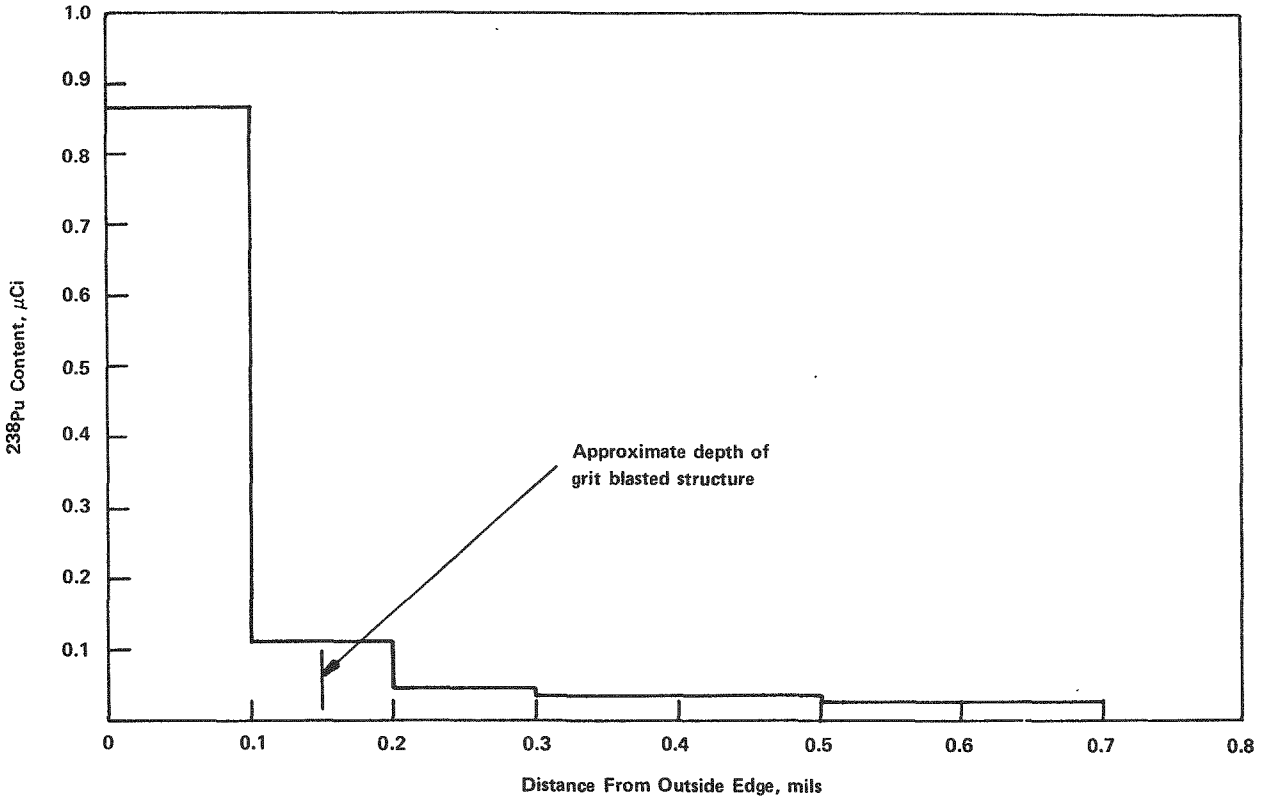

FIGURE 11 - Distribution of plutonium in the outside portion of the PISA wall. The amount of plutonium corresponds to the alpha radiation emitted from the electrolyte used to remove the "layers" from 0.11 in.^{2} of the surface.

The concentration of plutonium in the grit blasted structure and the fact that the PISA did not develop any leaks dictates that the plutonium originally came from a source outside the PISA, rather than from within it. This original source is the plutonium environment in which the PISA is fabricated. Subsequent acid decontamination does not remove the plutonium which is deeply embedded within the grit blasted structure and this plutonium effuses out during heating causing the observed surface contamination.

Conclusions

Acid decontamination of plutonium from the exterior surface of the PISA effectively removes the superficial plutonium but does not extract the plutonium which is deep within the grit blasted structure on the capsule wall and leaves roughly 100 μCi. Subsequent heating of the capsule at service temperature of 1350°C results in effusion of the plutonium out of the grit blasted structure with swipe measurements as high as 400,000 DPM after 1535 hr. The plutonium does spread to the interior of the GIS enclosure but had not diffused to the outside of the GIS after 1535 hr at temperature.

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