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RESULTS OF ANALYSES PERFORMED ON CONCRETE CORES REMOVED FROM FLOORS AND D-RING WALLS OF THE TMI-2 REACTOR BUILDING

Charles V. McIsaac
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Prepared for the
U.S. Department of Energy
Three Mile Island Operations Office
Under DOE Contract No. DE-AC07-76ID01570

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CONTENTS

ABSTRACT

The March 28, 1979 loss-of-coolant accident at Three Mile Island Unit 2 (TMI-2) exposed about 7,200 m^2 of concrete surfaces within the Reactor Building to liquid and vapor-phase contaminants. The majority of those surfaces are protected by coatings of epoxy-based, nuclear grade paints. During September 1983, seventeen high quality cores were extracted from the concrete floors and D-ring walls at the 305- and 347-ft elevations of the Reactor Building. The samples were subjected to a series of analyses to determine the surface concentrations of radionuclides present and to characterize their distribution in the coatings layer and sub-surface concrete.

The analysis results indicate the protective coatings provided significant protection against radionuclide penetration. Radiocesium, in most cases, was found to be confined within a few millimeters of the top surface of the coatings layer. However, at floor locations having coatings that were damaged prior to the accident, radiocesium penetrated into the sub-surface concrete to a depth of several centimeters. Parametric calculations performed using the ISOSHLD-2 computer code indicate that the decontamination of the 347-ft elevation floor will reduce the general area gamma exposure rates at this elevation by as much as 40%. The data indicate that scabbling would be an effective decontamination technique.

INTRODUCTION

During the first three days following the shutdown of the TMI-2 reactor, an estimated 1.0×10^6 L of contaminated primary coolant escaped from the primary system through the pressure-operated relief valve (PORV) on the pressurizer. This lost coolant flowed through interconnecting piping to the Reactor Coolant Drain Tank (RCDT), located in the south-west quadrant of the Reactor Building basement. The rupture disk on the RCDT burst within minutes of the initial opening of the PORV, allowing primary coolant, along with hydrogen gas generated by the metal/water reaction in the core, to pass from the tank through a vent line to the Reactor Building basement. Sufficient hydrogen was released to the Reactor Building atmosphere by this pathway to cause a detonation, which in turn actuated the post-LOCA containment spray system. This system, located on the Reactor Building dome, sprayed 6.4×10^4 L of water treated with boron and sodium hydroxide into the Reactor Building atmosphere. During this time, the Reactor Building air cooling assembly on the 305-ft elevation floor was circulating moisture-laden air from its immediate environment to the Reactor Building dome.

The Reactor Building atmosphere likely remained saturated with water for months following the accident, resulting in extensive pooling of contaminated water on upper-level horizontal surfaces. With the exception of the Reactor Building basement impingement walls, all Reactor Building concrete surfaces are protected from contamination penetration by coatings of epoxy-based, nuclear-grade paints supplied by Keeler and Long, Inc. The coatings provide a resilient, water resistant surface for the otherwise porous concrete.

The first systematic sampling of Reactor Building surfaces took place during December 1981 and March 1982, before and after the Reactor Building gross decontamination experiment.[1] Surface samples were collected using a milling tool developed by EG&G Idaho, Inc. One of the objectives of the surface sampling program was to determine the depth of penetration of activity into structural concrete. However, because of the nature of the sampling method used, the results were inconclusive. Therefore, a second

sampling campaign that used a different sampling technique was conducted during September 1983. Seventeen core samples, each approximately 4.5 cm in diameter by 5.5 cm in length, were removed from Reactor Building concrete floors and D-ring walls.

Following their removal from the Reactor Building, the core samples were subjected to analyses under the general guidance found in Reference 2. Prior to any destructive analyses, the beta and gamma exposure rates on the top and bottom surfaces of each core were measured using two types of radiological survey instruments and thermoluminescent dosimeters (TLDs). The whole cores were initially analyzed using gamma ray spectroscopy and autoradiography analysis techniques. Several of the cores were then longitudinally sectioned and one section from each was cut into wafers. These wafers were individually analyzed for activity content. Coatings removal experiments were performed on longitudinal sections cut from four cores. In addition, six samples, including both sectioned and whole core samples, were longitudinally gamma scanned using a highly collimated intrinsic germanium spectrometer.

The autoradiography and whole core gamma spectroscopy analysis results are reported in Reference 3. This report presents those findings and, in addition, presents the results of the longitudinal gamma scans and the concrete wafer analyses.

SAMPLING AND MEASUREMENT METHODS

Sampling Locations

The total surface area of concrete floors, walls, and other structural components within the Reactor Building is about 7,200 m^2. Since only a modest number of core samples were to be collected, the selection of sample locations was primarily based upon the following two criteria; (a) sample locations must be representative of the major concrete surfaces at each accessible elevation and some must be in areas where contaminated water pooled for long periods of time and (b) a limited number of samples must be collected at locations that exhibit visible surface scars or other signs of damage to the coatings layer. Based upon the above criteria and visual inspections of Reactor Building concrete surfaces, 17 sampling locations were chosen. The locations selected are shown in Figures 1 and 2, which are, respectively, the floor plans of the 305- and 347-ft elevations. The sampling locations are indicated in these figures by darkened circles and corresponding sample numbers. Locations 305-2, 305-7, and 347-3 are on the 'B' D-ring wall; all other locations are on either the 305- or 347-ft elevation floors.

Core Boring

Concrete core samples were collected using a Drillco Flex-Shaft core boring machine equipped with a 5.08-cm outside diameter core-boring bit with a diamond surface. All drilling was conducted at 800 rpm using a slow bit advance rate. In order to avoid leaching the concrete during core removal, liquid bit coolant was not used. Adequate cooling was achieved by forcing a small quantity of compressed air though the hollow bit during boring. To maintain good cutting quality and to prevent sample cross contamination, the bit was changed after each drilling. The concrete cores were typically 4.5 cm in diameter and 5.5 cm in length.

At the sampling site, each core was sealed inside a clean plastic bag. All samples were later transferred to a clean, plastic-lined glovebox where they were visually inspected and cleaned of the loose dust which had

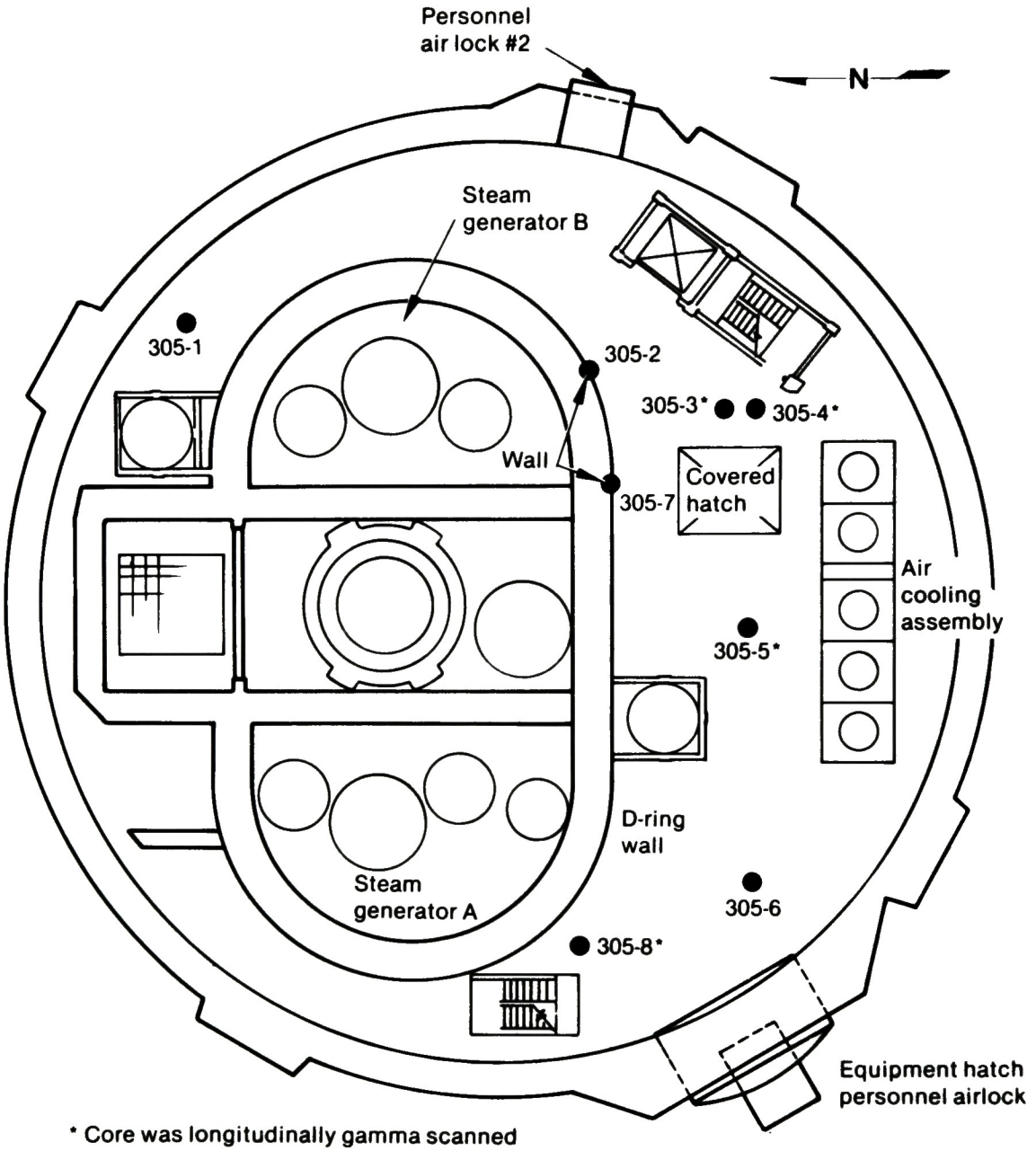
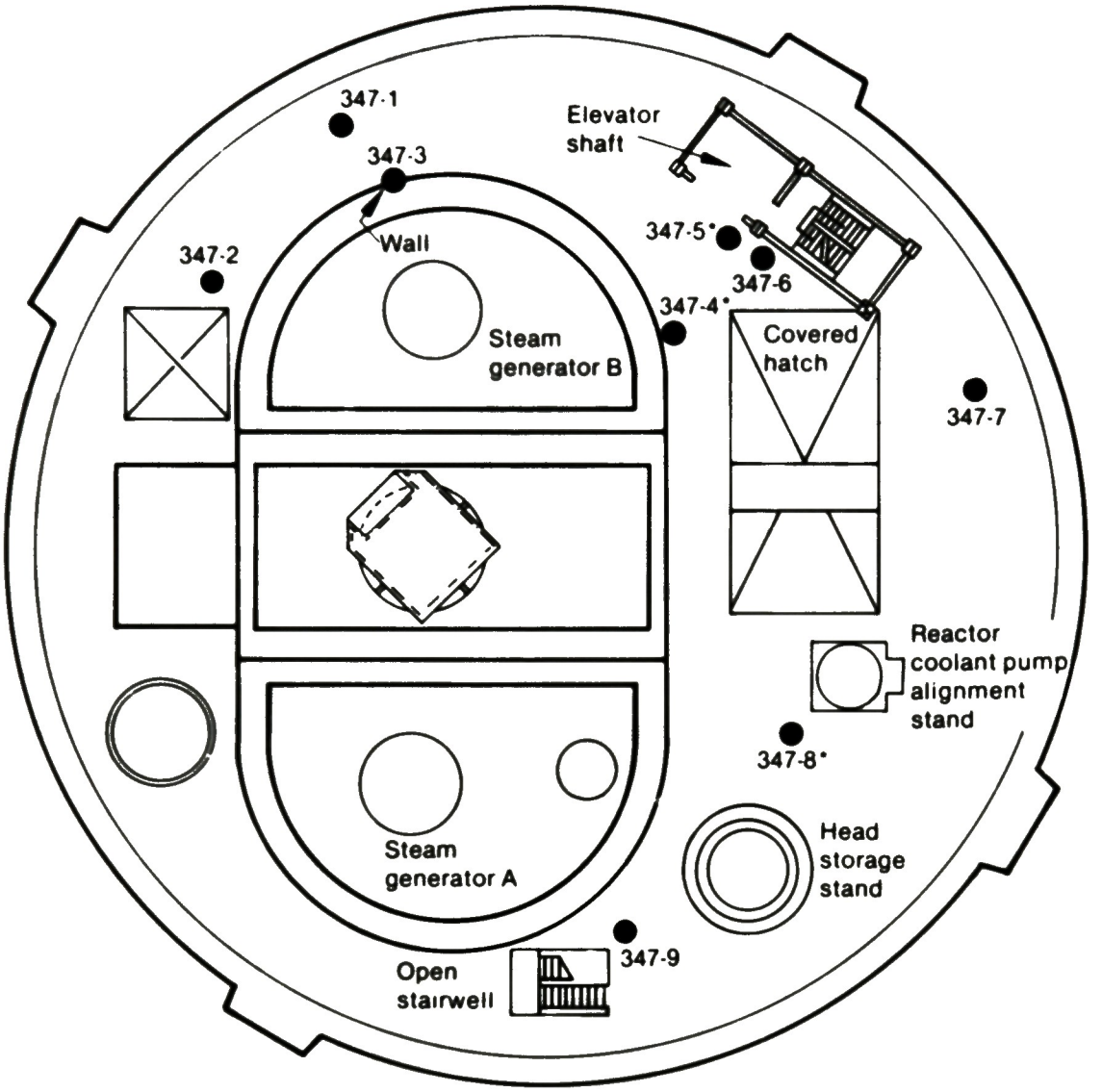


Figure 1. Locations at the 305-ft elevation where concrete cores were removed.



* Core was longitudinally gamma scanned

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Figure 2. Locations at the 347-ft elevation where concrete cores were removed.

accumulated during drilling. Each sample was wiped gently with a dry cloth until no visible dust was transferred from the sample to the cloth. After cleaning, each core was transferred to a clean polyethylene bag and placed inside a second polyethylene bag to provide additional protection against the release of contamination.

Exposure Rate Measurements

Following the cleaning described in the previous section, each core was subjected to beta and gamma exposure rate measurements while still enclosed in the two polyethylene bags. Exposure rate measurements were made at both the top and bottom surfaces of each core using an Eberline RO-2 ionization chamber survey meter and an Eberline E-530N Geiger-Muller survey meter that was equipped with a shielded probe having a certified 50 to 1 front-to-back ratio. In addition, measurements were made using Panasonic TLDs that had windows that were 7, 300, and 1000 mg/cm^2 thick. The TLD measurements were all made while the plane of the TLD chips was located 2.54 cm away from the painted surface of each core. The average TLD exposure time was about 47 hours. The TLDs were read using the on-island GPU Nuclear TLD reading system.

Whole Core Gamma Ray Spectroscopy

The inventory of gamma-ray-emitting radionuclides in each sample core was measured using GPU Nuclear lithium-drifted germanium [Ge(Li)] spectrometers. During each measurement, the painted circular surface of the core was oriented towards the detector and the counting geometry was the same as that used to assay 5.08-cm diameter particulate filters. This counting geometry was chosen so that the photopeak count rate data obtained could be converted to radionuclide concentrations using available counting efficiency tables. The counting efficiency tables for the particulate filters that were used were not corrected for sample self-absorption; but calculations have been performed that show that for activity evenly distributed to a depth of 1.27 cm within a concrete core having the same dimensions as the samples, the correction is only about 25%.[4]

Autoradiography

Autoradiography is a measurement technique that involves placing a radioactive source in intimate contact with a photographic film to produce a latent image of the distribution of activity across the source. The sample cores were autoradiographed by placing the painted surfaces on 10.16- by 12.7-cm Polaroid Land ASA 400 speed film for periods of time ranging from 30 minutes to 24 hours. Many cores were also autoradiographed on Kodak Xomat AR film for comparison. Figure 3 shows the autoradiograph of sample number 305-7 overlaid on a photograph of the painted surface of the sample so that a comparison may be made between surface features and activity distribution.

Sample Sectioning and Leaching

Five cores were longitudinally sectioned using a Buhler lapidary saw equipped with a 0.89-mm thick diamond tipped blade. To minimize the spread of contamination, no coolant was used to cool the blade and cutting was directed from the bottom of each core towards the painted surface. Four of these longitudinal sections were cut into wafers, each wafer being about 8 mm thick. Three such wafers were obtained from the painted end of each of the four core sections.

The twelve wafers thus obtained were analyzed for activity content using an acid leach technique. Each wafer was sequentially immersed in concentrated hydrochloric acid baths until discoloration of the acid ceased. The acid leachants and a sample of the concrete residue were then analyzed for gamma-ray-emitting radionuclides using the on-island Pacific Northwest Laboratory (PNL) Ge(Li) spectrometer system.

In order to determine the efficacy of paint removal as a decontamination method, the coatings on the surfaces of four core sections were removed. The process was completed in four stages on two of the samples and in two stages on the remaining two samples. After each stage, the core section was counted for 5 minutes with a Ludlum model

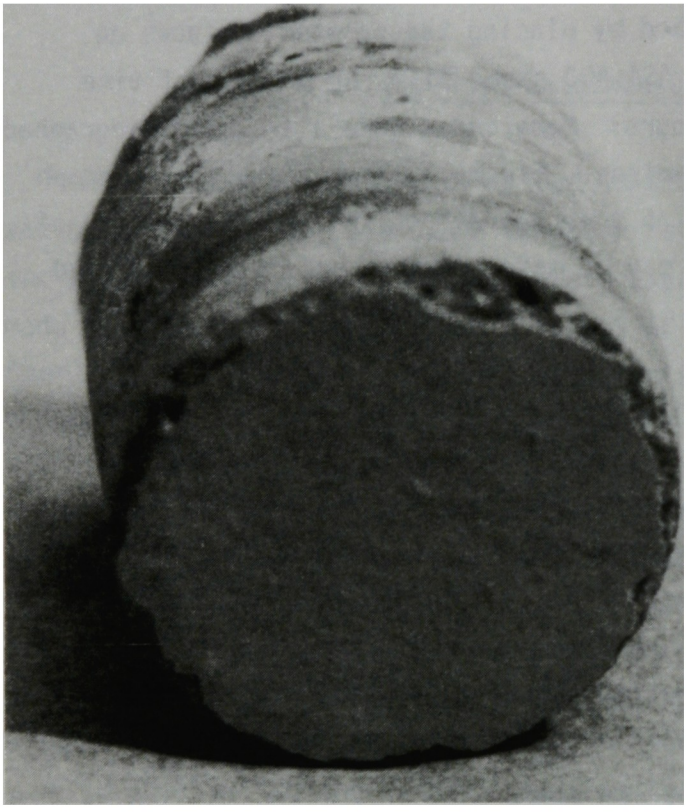
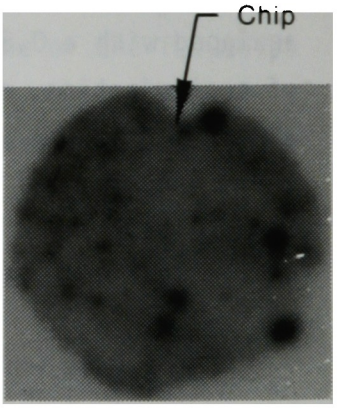


Photo of
core number
305-7



Autoradiograph
exposure

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Figure 3. Concrete core sample autoradiograph showing dark shading where the coating protected the concrete from radiation penetration.

2000 Geiger-Muller scaler using a 5.08-cm diameter end window (1.5 to 2.0 mg/cm^2 mica window) detector. The sample-to-detector counting distances used were 5 and 2 cm for each core.

The first three steps in the coatings removal process were sequential sandings of the painted surface with 400- and 800-grit sandpaper. The coating consists of three layers, which, in the order that they were originally applied on the concrete surfaces, are Keeler and Long No. 6548 epoxy block filler, No. 7107 epoxy white primer, and No. 7475 epoxy white enamel finish paint. Keeler and Long[5] reports the dry film thicknesses of those paints are, respectively, 0.142, 0.076, and 0.064 mm. The initial sanding restored the coatings original color, that being cream white. The second step was a repeated light sanding. The third sanding probably removed the majority of the finish and primer layers. The fourth and final step in the coatings removal process used a Dremel electric hand tool with fine stones that is normally used to carve wood and soft stone. Following this last step, the surface was free of any coating except that in the inaccessible crevices and pores of the concrete.

During the course of removing the coatings, precautions were taken to prevent sample recontamination. Following each treatment, the core sample was vacuumed and tape was used to remove loose surface contaminants.

Collimated Source Gamma Ray Spectroscopy Measurements

To determine the distribution of radiocesium that had migrated through the 0.282 mm-thick coatings layer and into the sub-surface concrete, six core samples were longitudinally gamma scanned using a highly collimated intrinsic germanium spectrometer. The technique involved making repeated measurements of the count rate of the 662-keV photopeak of ^{137}Cs while moving the sample incrementally across the field of view of the spectrometer. A plot of count rate versus scan position provides a knowledge of the distribution of ^{137}Cs as a function of depth within a core (see Figures 4 through 9).

Each core was scanned while held in a bracket that kept the longitudinal axis of the core perpendicular to the vertical centerline of

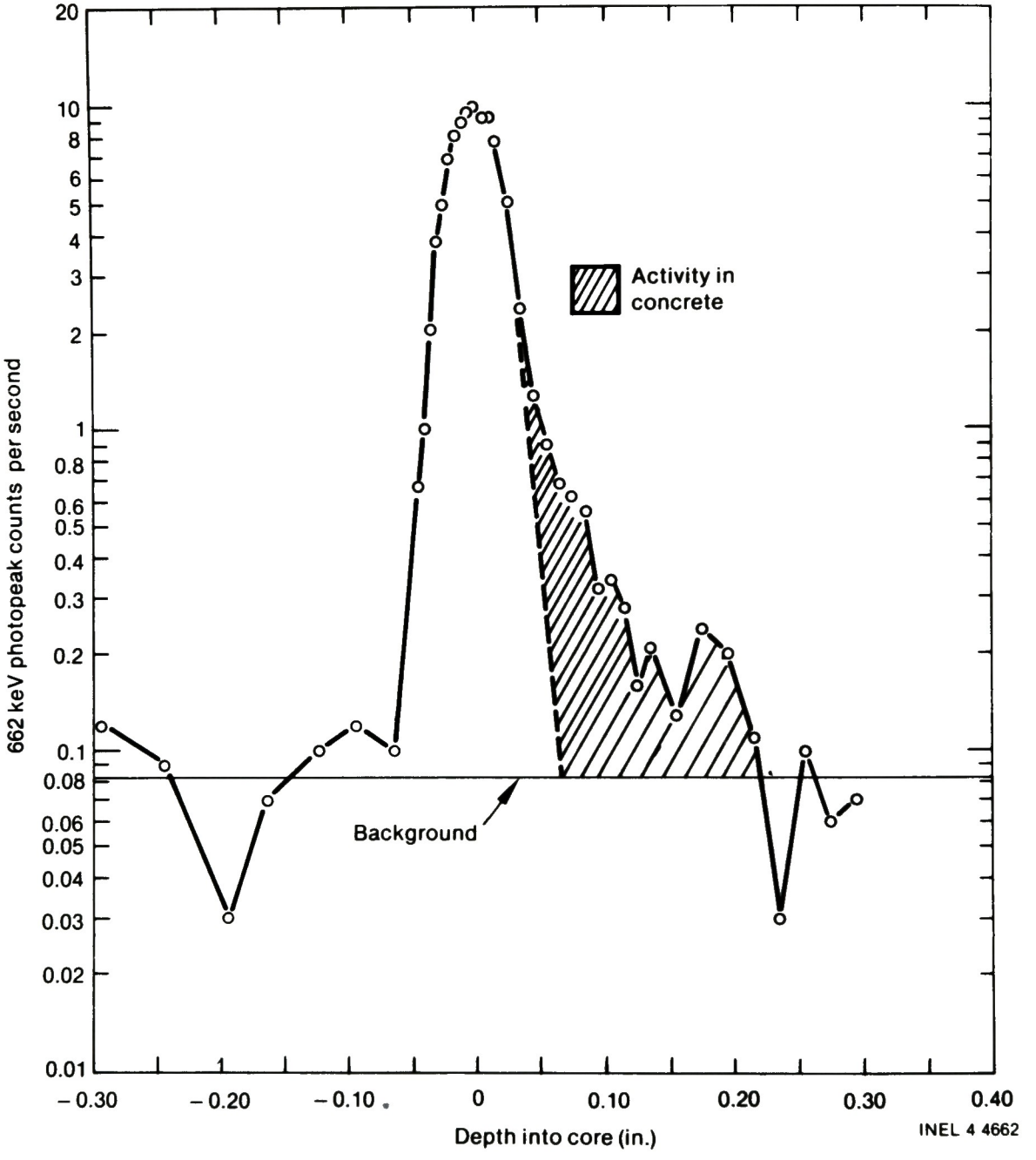

Figure 4. ^{137}Cs distribution profile within concrete core sample number 305-3.

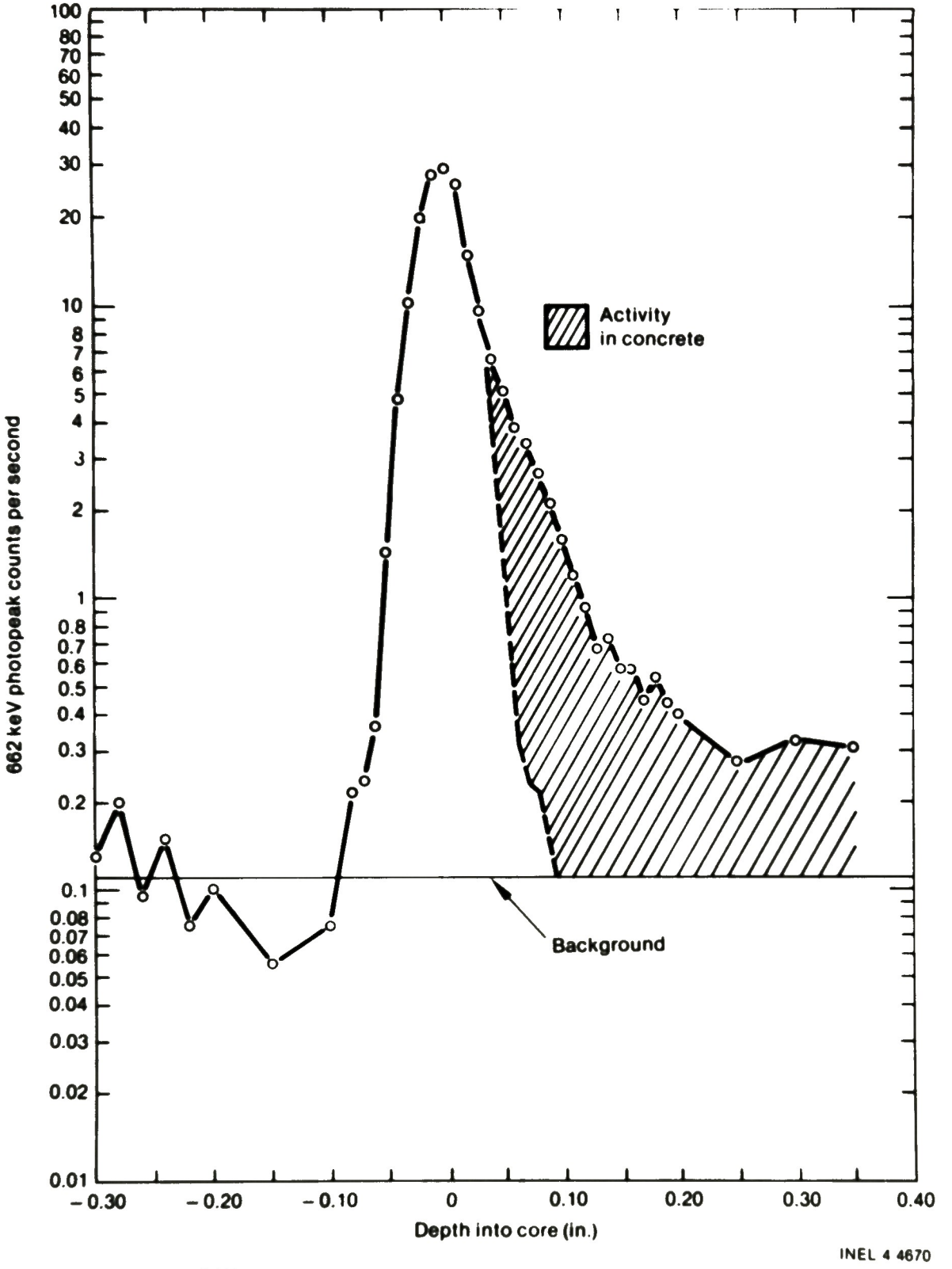

Figure 5. ^{137}Cs distribution profile within concrete core sample number 305-4.

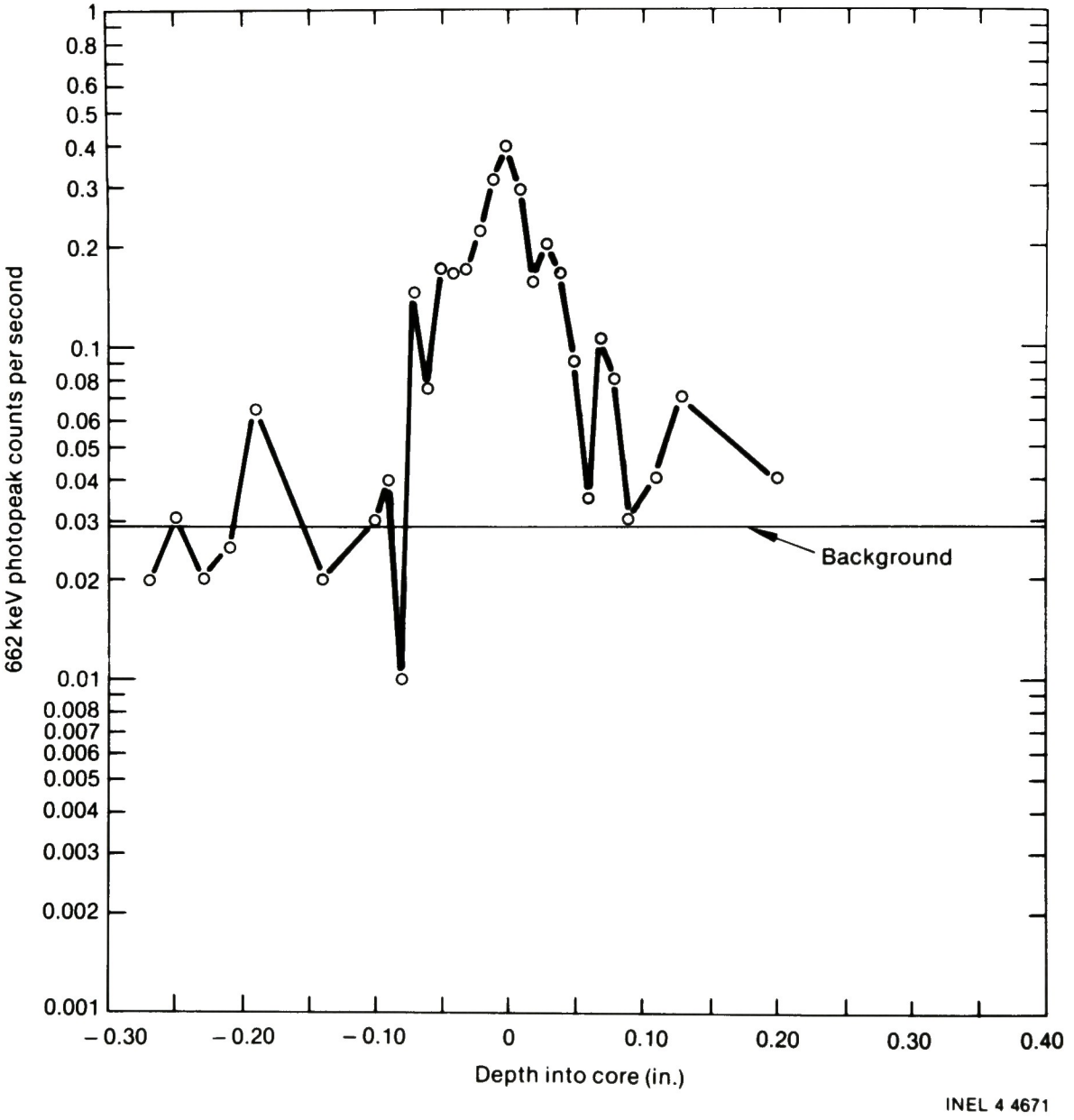

Figure 6. ^{137}Cs distribution profile within concrete core sample number 305-5.

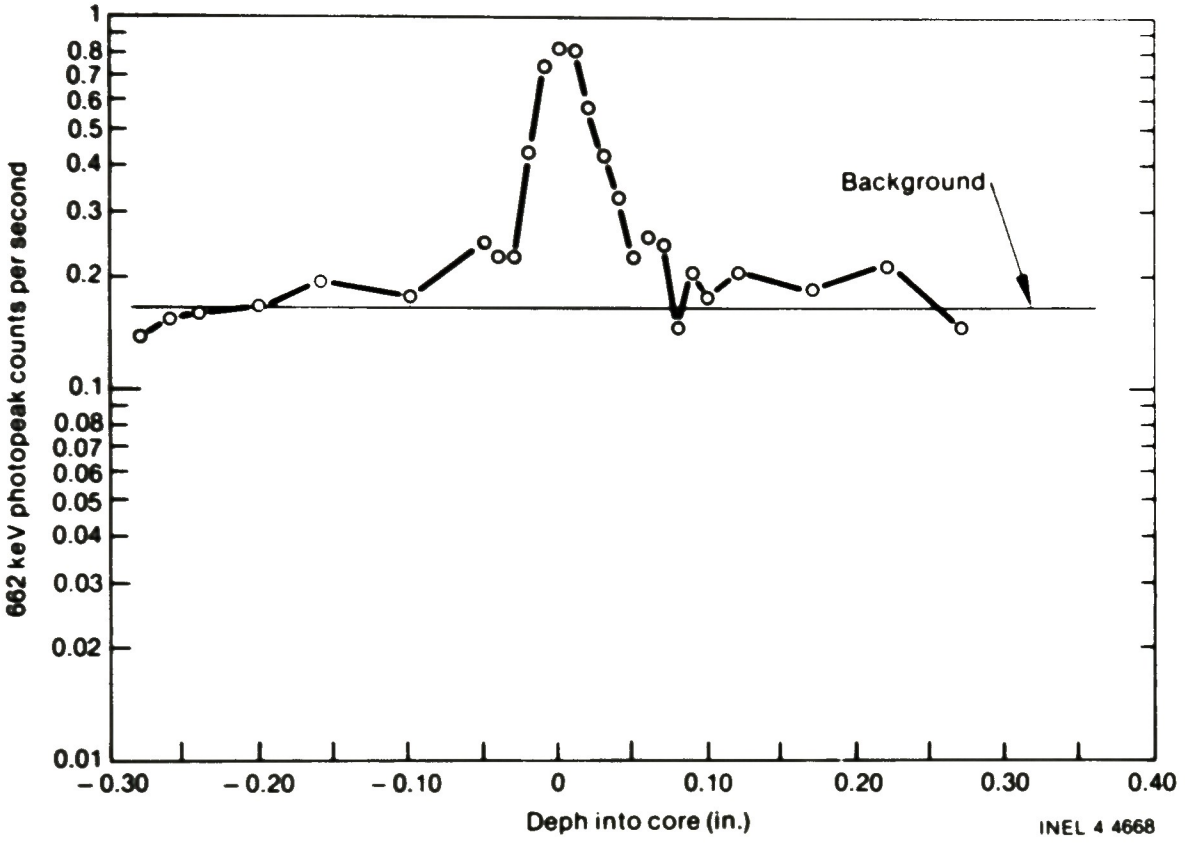

Figure 7. ^{137}Cs distribution profile within concrete core sample number 305-8.

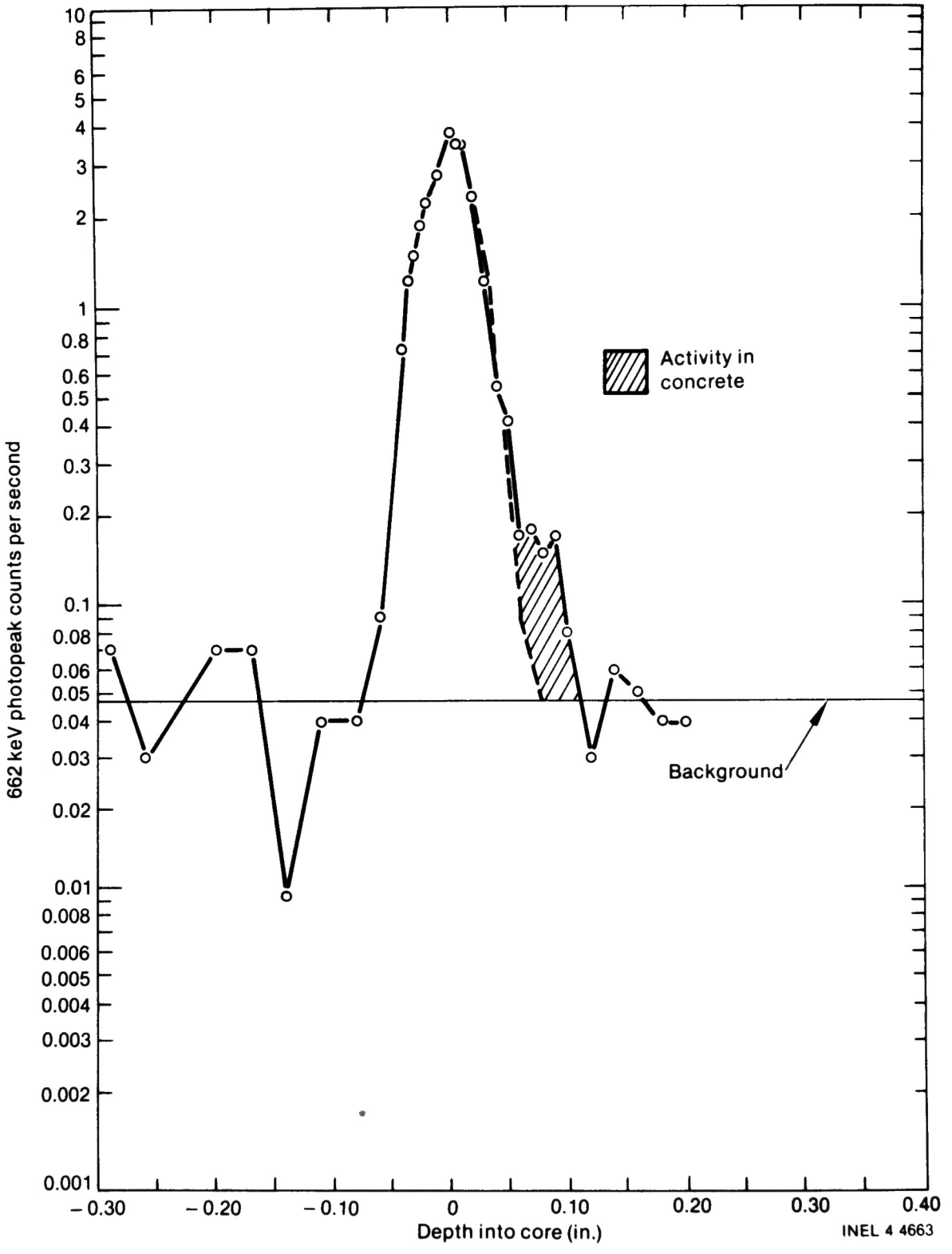

Figure 8. ^{137}Cs distribution profile within concrete core sample number 347-5.

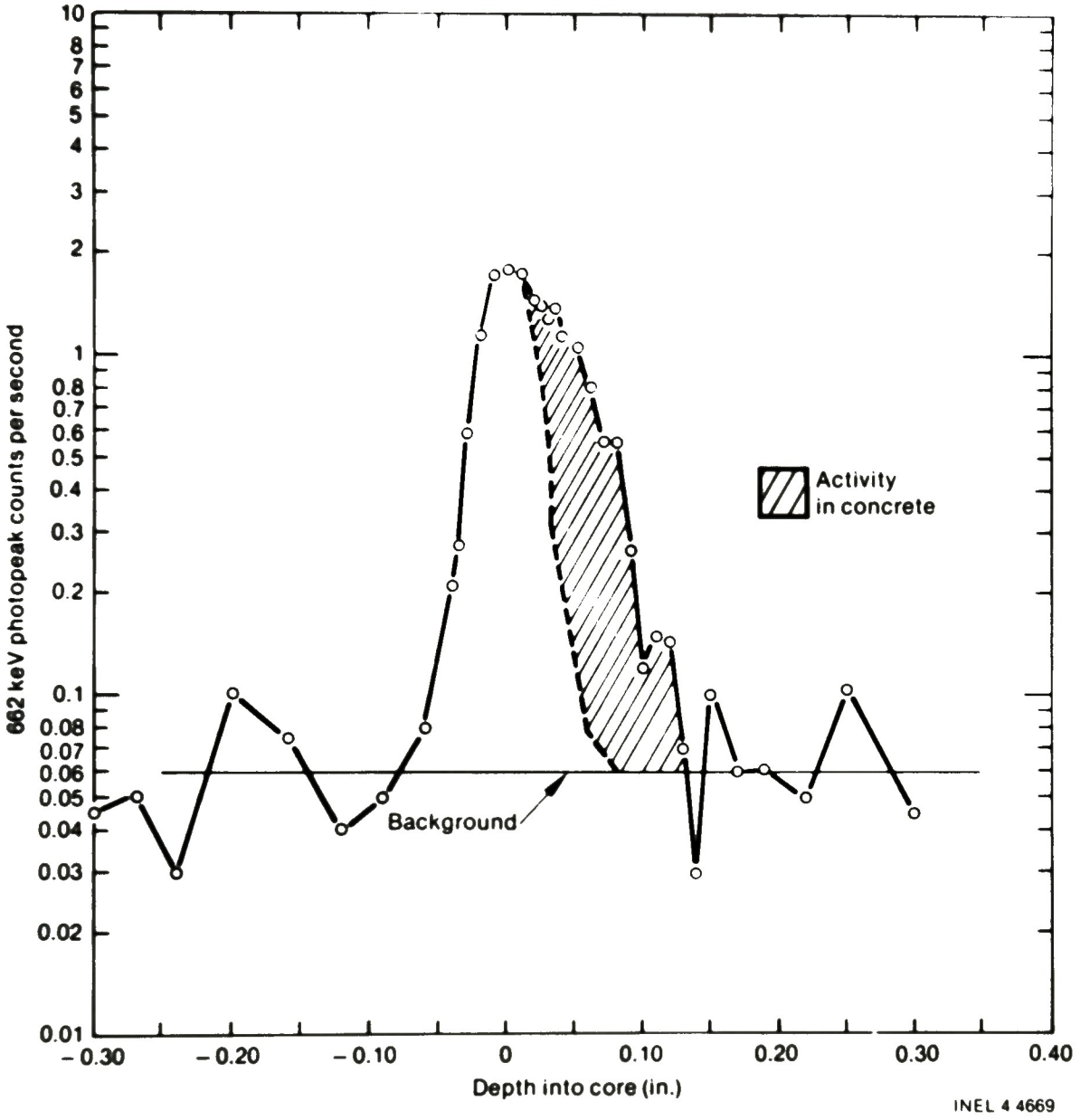

Figure 9. ^{137}Cs distribution profile within concrete core sample number 347-8.

the collimator's aperture. The aperture, which measured 20.3 cm high by 10.2 cm deep by 0.61 mm wide, was fashioned by sandwiching thin shims between two lead bricks whose surfaces had been machined flat to within a tolerance of $\pm$ 0.025 mm. During counting, the detector's end-cap was centered on the aperture and was fixed at a distance of about 3 mm from the rear surface of the lead bricks that formed the collimator. Each core sample was scanned while positioned so that the side of the core nearest the detector was at a distance of 2 cm from the front surface of the collimator. The cores were each initially positioned so that the painted surface of the core was conservatively outside the field of view of the detector. The painted surface was then moved towards the aperture in increments that ranged between 0.508 and 1.27 mm. The use of these relatively large steps continued until a significant increase in count rate was detected, at which time the increment was reduced to 0.127 or 0.254 mm. In each case, the average count rate measured while the surface of the core was outside the detector's field of view, was considered to be the background count rate for that sample.

In order to determine the resolution of the collimator, a contaminated particulates filter was gamma scanned using the same procedure used to scan the core samples. Particulates were deposited on the filter over a region about 1.2 cm in diameter and the average surface density of the material was about 4 mg/cm^2. The thickness of the deposit was not measured, but for an assumed density of 3 g/cm^3 this surface density translates into an average thickness of 0.01 mm. Certain regions of the deposit may have been an order of magnitude thicker than this.

The filter was mounted on one end of an uncontaminated concrete core section during the measurements so as to duplicate the case where all of the activity is bound in the coatings layer and none resides in the concrete. The filter was scanned while its near edge was positioned at distances of 2 and 3 cm from the front face of the collimator. The activity profiles that were measured are shown in Figures 10 and 11.

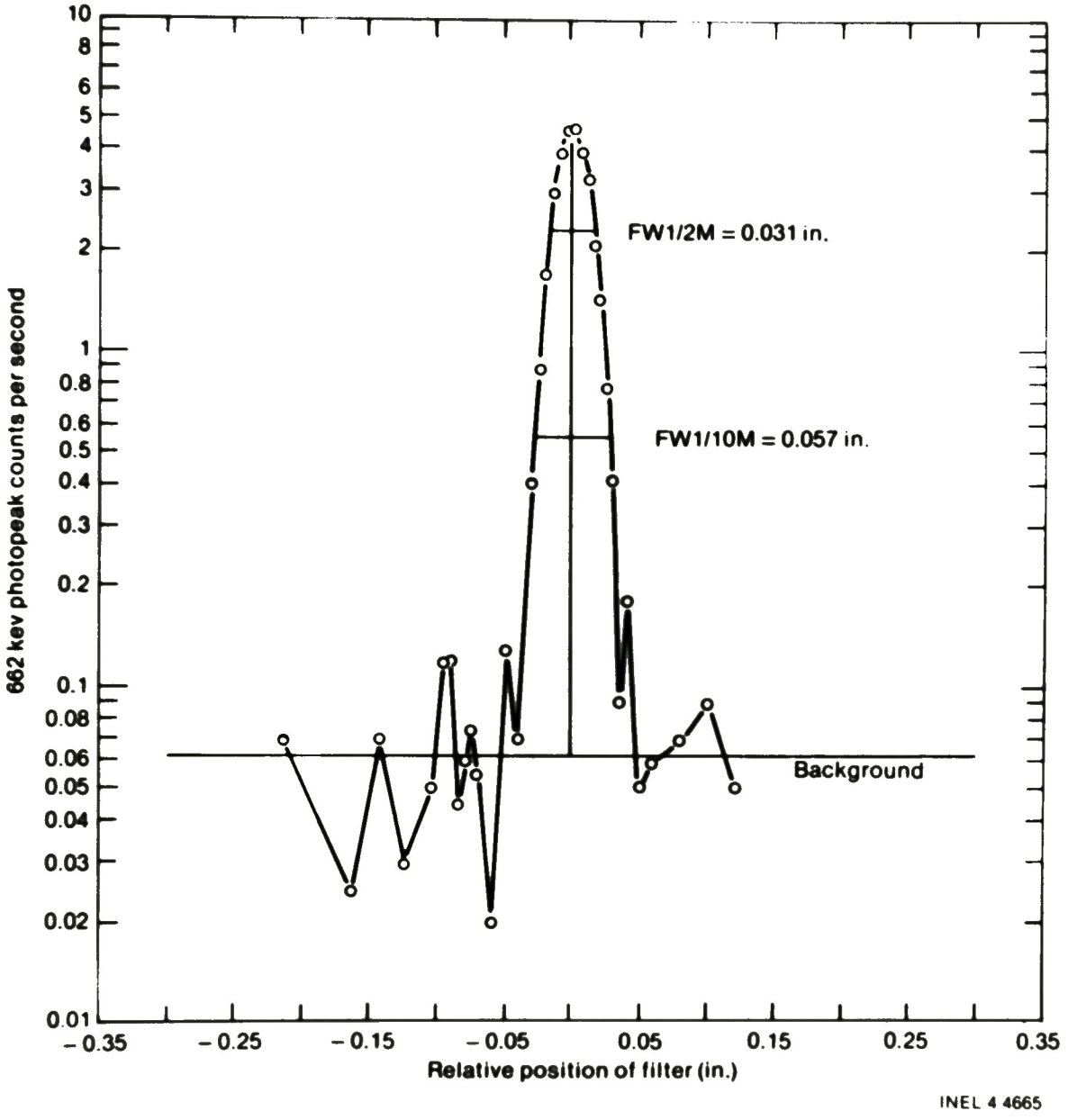

Figure 10. Measured ^{137}Cs activity profile of a contaminated particulates filter that was gamma scanned at a distance of 2 cm.

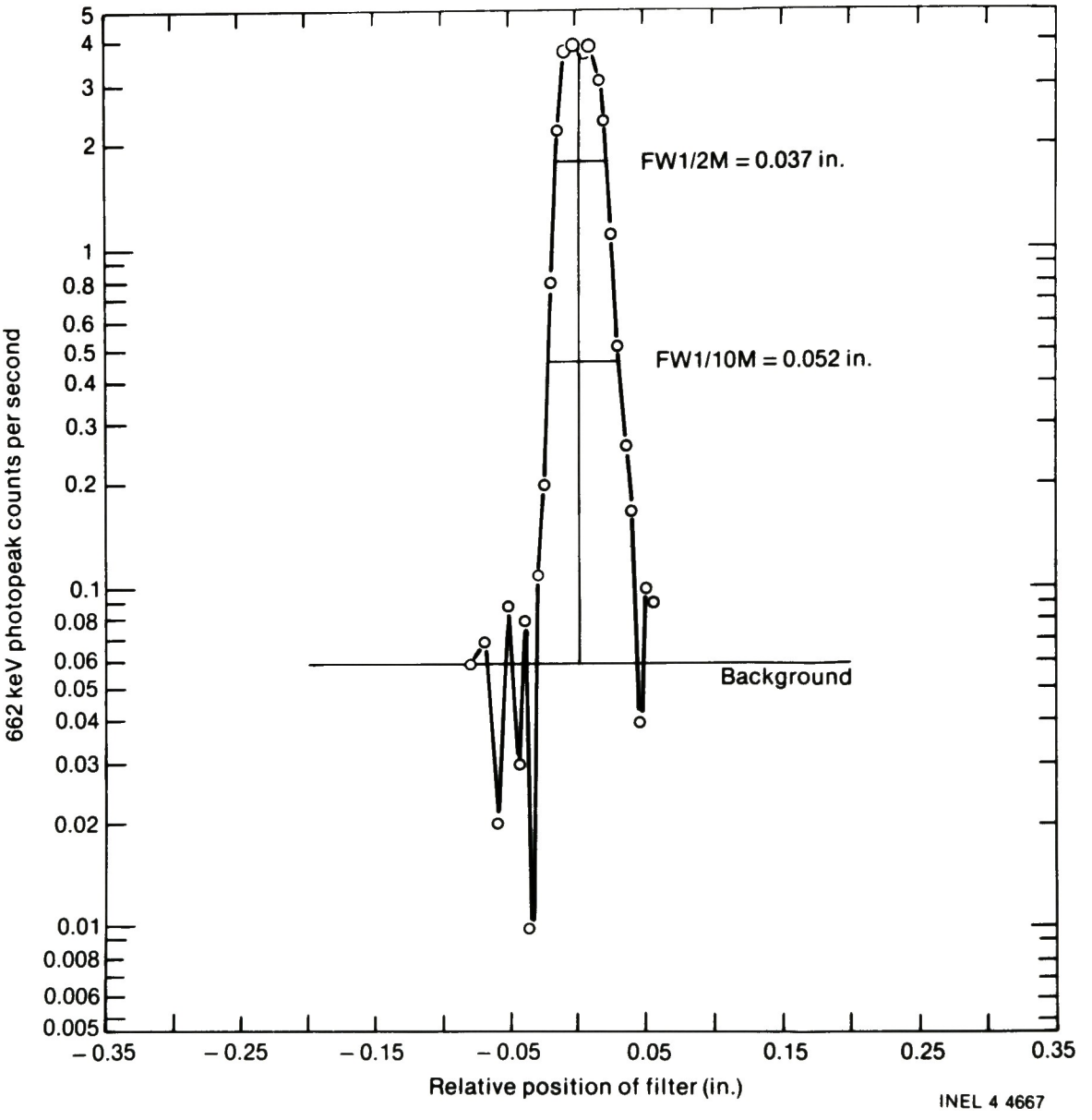

Figure 11. Measured ^{137}Cs activity profile of a contaminated particulates filter that was gamma scanned at a distance of 3 cm.

DISCUSSION OF RESULTS

Radiocesium Surface Concentrations

The surface concentrations of ^{134}Cs and ^{137}Cs that were measured by gamma ray spectroscopy prior to the start of any destructive analyses on the core samples are presented[3] in Table 1. The concentrations are equivalent surface concentrations since they were calculated by dividing the total activity measured by the core's cross-sectional area (i.e. 14.53 cm^2). The listed concentrations are decay-corrected to November 1, 1983.

The D-ring wall samples exhibit radiocesium surface concentrations that are on the average two orders of magnitude lower than those measured on floor samples. Cesium-137 concentrations on floor surfaces range from 0.011 $\mu Ci/cm^2$ at sampling location 347-7 to 5.3 $\mu Ci/cm^2$ at sampling location 305-3. The core sample having the highest surface activity concentration, sample 305-4, was too radioactive to be counted so it is only possible to estimate its ^{137}Cs surface concentration based on its gamma exposure rate. Using the results of the linear regression analysis that is described in a following section of this report, the ^{137}Cs surface concentration at location 305-4 was estimated to be about 16 $\mu Ci/cm^2$. (The 1000 mg/cm^2 TLD data was used to make this estimate.) This abnormally high ^{137}Cs surface concentration is, in part, due to the presence of a significant amount of subsurface activity at this location. The sample exhibited a deep scar in its protective coating that was of pre-accident origin which allowed activity to penetrate into the concrete in the near vicinity of the scar (See Figure 5).

The ratio of ^{137}Cs to ^{134}Cs concentrations as of November 1, 1983 is nearly constant at 16 to 1. This is consistent with computer projections of the core inventory of these two nuclides and with analysis results for other types of samples collected from the Reactor Building.

TABLE 1. EQUIVALENT SURFACE CONCENTRATIONS OF ^{134}CS AND ^{137}CS MEASURED IN WHOLE CONCRETE CORE SAMPLES USING GAMMA RAY SPECTROPHOTOMETRY[a]

Core Sample Number	^{134}Cs ($\mu Ci/cm^2$)[b]	^{137}Cs ($\mu Ci/cm^2$)[b]
305-1	8.3 E-3	1.4 E-1
305-3	3.2 E-1	5.3 E+0
305-4	--[c]	--[c]
305-5	8.9 E-3	1.4 E-1
305-6	7.6 E-3	1.2 E-1
305-8	1.4 E-2	2.3 E-1
305-2[d]	7.6 E-5	6.5 E-4
305-7[d]	1.4 E-3	2.4 E-2
347-1	6.5 E-3	1.1 E-1
347-2	5.4 E-3	8.9 E-2
347-4	1.8 E-1	2.9 E+0
347-5	6.9 E-2	1.2 E+0
347-6	4.2 E-2	6.7 E-1
347-7	7.6 E-4	1.1 E-2
347-8	5.2 E-2	8.3 E-1
347-9	1.7 E-2	2.8 E-1
347-3[d]	3.3 E-4	5.0 E-3

a. Activities as of 11/01/83.

b. Equivalent surface activities were calculated using a whole core surface area of 14.53 cm^2.

c. This sample was not measured because activity was too high.

d. This sample was collected from a D-ring wall.

A comparison between the mean ^{137}Cs surface concentrations that were determined by measurements made on the core samples and those that were determined by measurements made on samples collected a year and a half earlier during March 1982 using a surface milling technique, is presented in Table 2. (The concentrations listed in the table were all decay-corrected to a common date, November 1, 1983.) The mean ^{137}Cs surface concentrations that were determined using these two sampling techniques agree reasonably well. The large uncertainties in the mean values reflect the fact that local ^{137}Cs surface concentrations on a given generic surface varied over a wide range of values. Samples collected in the vicinities of floor drains, for example, exhibited activity concentrations that were an order of magnitude higher than those measured at locations where long-term pooling of contaminants did not occur.

The autoradiographs made of the painted surfaces of the cores reveal that the surfaces sampled contain a fairly uniform background of radionuclides trapped within the coatings layer that is dotted with small localizations of higher activity. In general, rust-colored spots and stains correspond to areas having the highest intensities in the autoradiographs. (See Figure 3 for a comparison of visual surface features with autoradiographic features of core sample number 305-7).

The present general area gamma exposure rate on the 347-ft elevation of the Reactor Building is about 100 mR/h. To estimate what fraction of this field is caused by contaminants trapped in the floor, parametric calculations were performed using the ISOSHLD-2 computer code.[4] Disk sources, which ranged in diameter from 3.04 to 9.14 m, were modeled to duplicate the floor surface source term. Based on a ^{137}Cs surface concentration equal to the average value measured to be present on the 347-ft elevation floor (0.76 $\mu Ci/cm^2$), the gamma exposure rate was calculated at a point 1 m above the center of each disk. The results are presented below.

Disk Diameter (m)	Exposure Rate (mR/h)
3.04	23
4.57	30
6.09	36
9.14	43

These data indicate that from about 20 to 40% of the current general area exposure rate at this elevation is due to contaminants trapped in the floor.

Beta and Gamma Exposure Rates

The beta and gamma exposure rates that were measured on the painted surface of each concrete core sample are presented[3] in Table 3. One of the primary motives for performing these measurements was to determine how well the exposure rates correlated with measured surface activity. If it could be shown that the exposure rates immediately above a surface were simply related to surface activity concentrations on that surface then it would be possible to infer surface activity concentrations on surfaces not yet sampled using the results of a relatively quick and simply survey measurement. Since ^{137}Cs was determined to be the predominant radionuclide on Reactor Building surfaces it was chosen as the nuclide having the best potential for being simply related to the beta and gamma exposure rates.

Least-squares fits of the data to the linear equation, $y = a + bx$, where $y = \mu Ci/cm^2$ of ^{137}Cs and x = beta (mrad/h) or gamma (mR/h) exposure rate, were performed. The coefficients of the equation were computed along with the coefficient of determination (r^2) separately using each of the 12 data sets listed in Table 3. The calculated coefficients are presented in Table 4 for 10 of these data sets. The results for the 300 mg/cm^2 TLD data are omitted since the TLD is not sensitive to the relatively low energy beta emitted by ^{137}Cs.

TABLE 2. COMPARISON OF MEAN ^{137}Cs SURFACE CONCENTRATIONS MEASURED IN MILLED SURFACE SAMPLES WITH THOSE MEASURED IN CONCRETE CORE SAMPLES[a]

Location	Milled Samples[b] ($\mu Ci/cm^2$)	Core Samples ($\mu Ci/cm^2$)	Ratio of Milled to Core Samples
Elevation 305 ft			
Concrete Floor	6.6 ± 9.6 E-1	1.2 ± 2.3 E+0	5.5 ± 13.0 E-1
D-ring Wall	2.0 ± 1.3 E-2	1.2 ± 1.7 E-2	1.7 ± 2.6 E+0
Elevation 347 ft			
Concrete Floor	2.8 ± 3.4 E-1	7.6 ± 9.6 E-1	3.7 ± 6.5 E-1
D-ring Wall	3.3 ± 2.4 E-2	5.0 E-3	6.6 E+0

a. Activities as of 11/01/83.

b. C. V. McIsaac, Surface Activity and Radiation Field Measurements of the TMI-2 Reactor Building Gross Decontamination Experiment, GEND-037, October 1983.

TABLE 3. EXPOSURE RATES MEASURED ON THE TOP SURFACE OF EACH CONCRETE CORE SAMPLE

Core Sample Number	Eberline RO-2		Eberline E530N	Panasonic TLD[a]		
				Beta (mrad/h)		Gamma (mR/h)
	Gamma (mR/h)	Beta (mrad/h)	Gamma (mR/h)	7 mg/cm^2	300 mg/cm^2	1000 mg/cm^2
305-1	1.0	94.0	2.5	131.03	1.84	0.97
305-2	0.2	--[b]	<1.0	0.89	0.00	0.21
305-3	32.0	1136.0	75.0	1814.92	52.40	31.69
305-4	60.0	1880.0	300.0	3528.42	35.28	93.81
305-5	1.0	78.0	5.5	140.29	0.65	0.77
305-6	1.0	94.0	2.5	169.15	1.39	0.81
305-7	0.2	5.0	<1.0	8.26	0.13	0.21
305-8	1.5	117.0	4.0	235.42	0.69	1.29
347-1	1.8	66.6	11.0	268.09	6.00	3.72
347-2	0.4	9.2	1.5	15.50	1.24	0.74
347-3	0.2	0.4	<1.0	0.91	0.00	0.22
347-4	6.0	300.0	45.0	877.78	39.33	25.56
347-5	3.2	94.0	27.5	391.40	15.08	0.74
347-6	2.0	96.0	11.0	269.57	8.09	4.02
347-7	0.2	3.2	<1.0	11.77	0.17	0.21
347-8	2.2	45.6	12.5	190.60	7.96	5.57
347-9	0.8	41.4	4.0	118.09	4.49	2.03

a. These numbers refer to the surface density of the TLD window used to discriminate energy.

b. Not measured.

TABLE 4. RESULTS OF LINEAR REGRESSION ANALYSIS

Survey Device	Coefficients[a] a	b	r^2	Type of Measurement
Eberline RO-2				
305-ft data	-0.027	0.1665	1.000	gamma
	-0.242	0.0049	0.996	beta
347-ft data	-0.227	0.4845	0.939	gamma
	-0.026	0.0096	0.914	beta
Eberline E530N				
305-ft data	-0.102	0.0720	0.999	gamma
347-ft data	-0.126	0.0619	0.917	gamma
Panasonic TLD				
305-ft data	-0.226	0.0030	0.991	beta (7 mg/cm^2)
	-0.010	0.1676	1.000	gamma (1000 mg/cm^2)
347-ft data	-0.093	0.0032	0.913	beta (7 mg/cm^2)
	+0.012	0.1140	0.972	gamma (1000 mg/cm^2)

a. Coefficients of linear equation, $y = a + bx$, where $y = \mu Ci/cm^2$ of ^{137}Cs and x = beta (mrad/h) or gamma (mR/h) exposure rate.

The resulting linear equations are plotted in Figures 12 and 13. Figure 12 plots the equations obtained using the beta exposure rate data while Figure 13 plots the equations obtained using the gamma exposure rate data. An examination of these graphs reveals that the beta and gamma exposure rates measured on the surfaces of samples collected from the 347-ft elevation using the Eberline RO-2 dose rate measurement instrument are biased low compared to the results obtained using the E530N and TLDs. The gamma exposure rates measured using E530N are higher by about a factor of two compared to the results obtained using the 1000 mg/cm^2 TLDs. However, if we ignore the 347-ft RO-2 data, predicted ^{137}Cs surface concentrations vary by only a factor of about 2 using the beta exposure rate linear equations and they vary by a factor of about 3 using the gamma exposure rate linear equations.

Because the exposure rate measurements were made in a low-background environment outside of the Reactor Building, the linear equations relating exposure rates to ^{137}Cs surface concentration that were presented in Table 4 must be used with some caution when predicting surface activity based on measurements made inside the Reactor Building. This is particularly germane when gamma exposure rate measurements are used since survey instruments are not often sufficiently collimated to provide an accurate reading of the exposure rate arising from a small region of a large surface source. Given this circumstance, the use of beta exposure rates to predict surface activities is likely to be a more reliable method than one that uses gamma exposure rates.

Depth of Radionuclide Penetration into Coatings and Concrete

Several different types of measurements were performed on various core samples in an effort to characterize the distribution of radionuclides within and below the coatings layer. They included a coatings removal experiment, autoradiography measurements, radiochemical analyses performed on wafers cut from core samples, and gamma ray spectroscopy measurements that used a highly collimated intrinsic germanium spectrometer.

The data obtained from the coatings removal experiment that was performed on longitudinal sections cut from four core samples are presented[3] in Tables 5 and 6. The results for samples 347-2 and 305-1 are expressed as

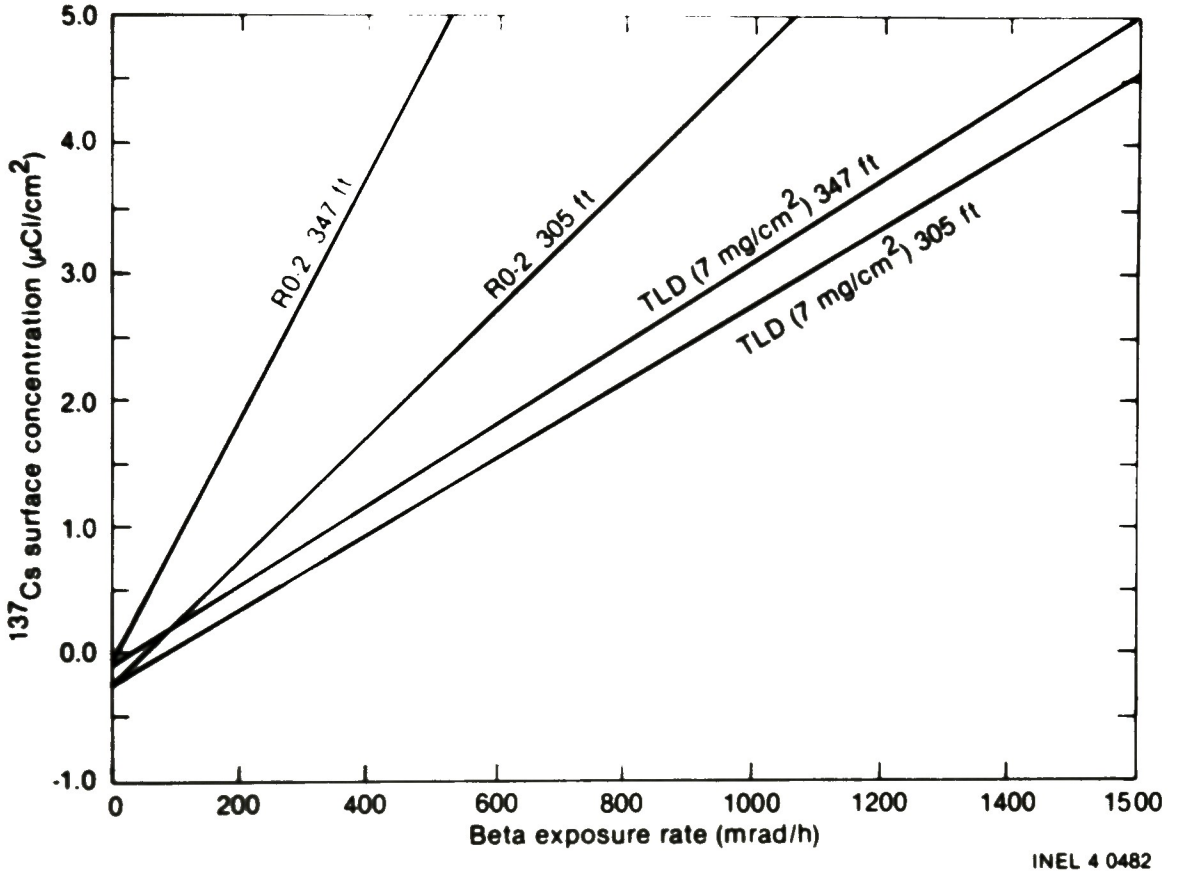

Figure 12. ^{137}Cs surface concentration versus beta exposure rate.

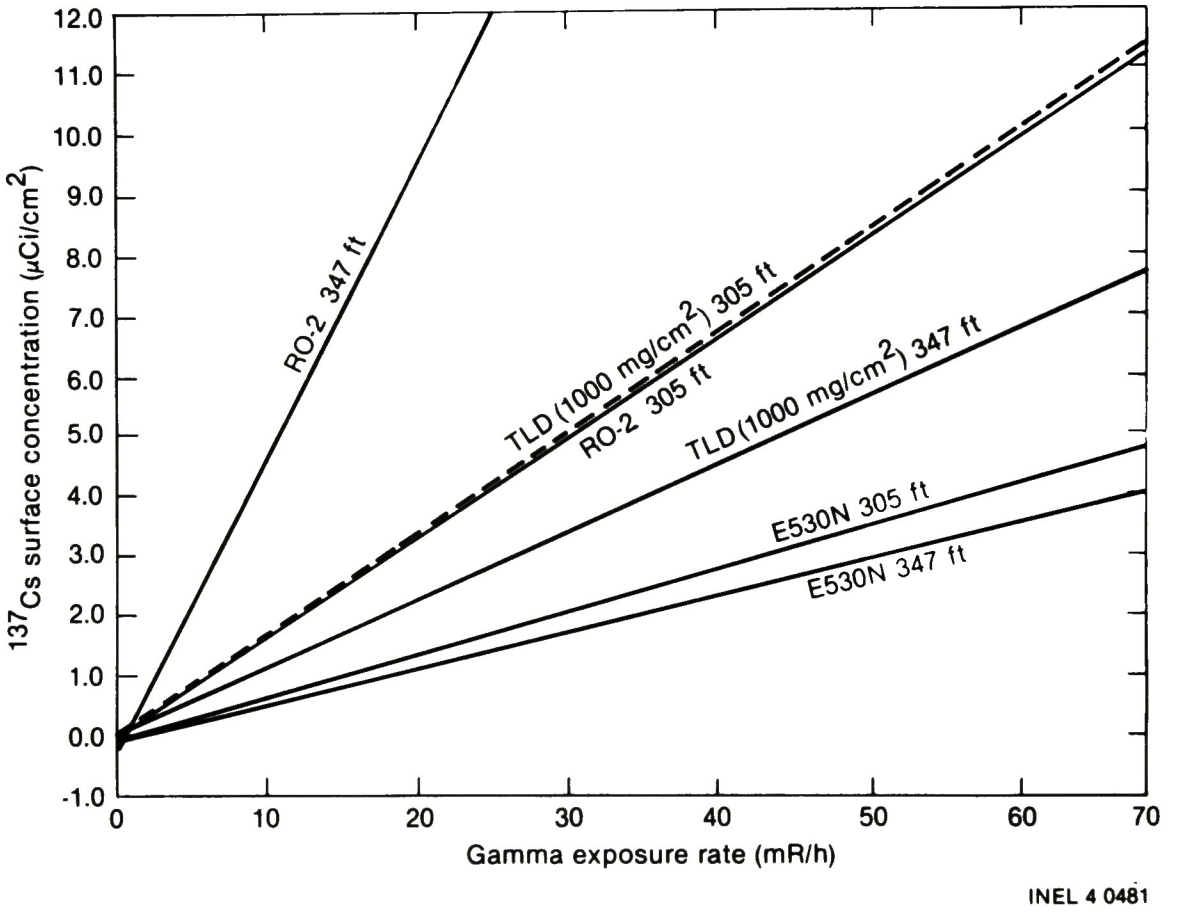

Figure 13. ^{137}Cs surface concentration versus gamma exposure rate.

TABLE 5. RESULTS OF COATINGS REMOVAL EXPRESSED AS PERCENT REDUCTION IN MEASURED DISINTEGRATIONS PER MINUTE

Core Sample Number	Coatings Removal Step			
	1	2	3	4
347-2	85.30	91.80	95.75	98.18
305-1	85.84	--[a]	--[a]	99.96

a. No intermediate steps were used to remove coatings layer.

TABLE 6. RESULTS OF COATINGS REMOVAL ON SURFACE EXPOSURE RATE

Core Sample Number	Initial		Final		Percent Reduction	
	mR/h	mrad/h	mR/h	mrad/h	mR/h	mrad/h
347-4	10	400	4	200	60	50
305-4	25	700	16	1000	36	-42

cumulative percent reduction in gross activity, measured in disintegrations per minute, following each stage in the coatings layer removal process. The data for sections 347-4 and 305-4 are expressed as percent reductions in beta and gamma exposure rates following removal of the paint from the top surfaces. The coatings on those latter two sections were removed in a single step.

The initial light sanding of core sections 347-2 and 305-1 reduced the measured gross activities of both sections by about 85%. The original color of the painted concrete surfaces within the Reactor Building had been a glossy cream white, however, it was generally observed that the color had yellowed. The cores collected from the floors exhibited considerably more yellowing than did those obtained from the D-ring wall. The yellowed layer was easily removed by light sanding and it is estimated that this layer is between 0.051 and 0.102 mm thick. Based on a coatings layer thickness of 0.282 mm, the maximum thickness of the yellowed layer is about one-third that of the coatings layer. Completely removing the coatings from sections 347-2 and 305-1 reduced their measured gross activities by 98.18 and 99.96%, respectively.

The removal of the coatings from sample sections 347-4 and 305-4 produced less dramatic results. The samples' gamma exposure rates were reduced by 60 and 36%, respectively. The beta exposure rate of sample 347-4 decreased by 50%, however, the beta exposure rate of sample 305-4 actually increased by 42%. This indicates that moderate quantities of beta-emitting nuclides migrated through the coatings layer at this latter location and that the coatings layer was acting as an effective beta shield. As was previously mentioned, sample 305-4 has a deep scar in its protective coating that is of pre-accident origin and the location at which the core was removed had been covered by standing water for an extended period of time.

The autoradiographs of sample 305-4 confirm the findings of the coating removal experiment. Autoradiographs of whole cores and longitudinal sections of cores that were made by laying the samples length-wise on photographic film, indicate that activity penetrated into

the concrete at only one other location, that being 305-3. The autoradiographs of these two samples indicate that the maximum depth activity penetrated is 2 mm at location 305-3 and 3 cm at location 305-4. To further investigate this phenomenon, longitudinal sections from those and two other samples were cut into wafers and subjected to radiochemical analyses.

The results of those analyses are presented in Table 7. The table presents the ^{137}Cs activities that were measured in the paint and the concrete wafers obtained from core sections 305-1, 305-3, 305-4, and 347-4. The paint that was removed from section 305-4 and 347-4 during the coatings removal experiment was, unfortunately, lost, so for those two samples only the activities that were measured in the wafers are presented. The depth intervals shown in the table correspond to the coatings layer (0.00 to 0.28 mm) and to the three wafers cut from successively deeper regions of each core section, each wafer being about 8 mm thick.

The data in Table 7 support the conclusion, first suggested by the autoradiography result, that activity penetrated into the concrete at sampling locations 305-3 and 305-4, but they also indicate that radiocesium penetration occurred at sampling locating 347-4. The autoradiograph of the top surface of sample 347-4 exhibited a high intensity region whose position and shape corresponded to a shallow groove that runs across one quadrant of the surface. As is suspected to be the case with sample 305-4, this type of surface feature may be responsible for the observed deep activity penetration.

The data presented in Table 7 for samples 305-1 and 305-3 support contradictory conclusions regarding the significance of penetrated activity. Essentially all (i.e. 99.87%) of the activity in core section 305-1 was found to be bound in the coatings layer but only 3.5% of the total activity measured in core section 305-3 was found to be associated with its coatings layer. In all cases, the majority of the penetrated activity was found to reside in the top-most wafer, or, in other words, within a depth of 8.2 mm from the coatings layer surface. The top wafers were found to contain from 88.3 to 99.9% of the total activity measured in each set of wafers. Within the depth interval 9.12 to 17.17 mm similar percentages range from 11.6% for sample 347-4 to 0.04% for sample 305-3.

TABLE 7. Cs-137 ACTIVITIES MEASURED IN PAINT REMOVED AND CONCRETE WAFERS CUT FROM SELECTED CONCRETE CORE SAMPLES[a]

Depth Interval (mm)	Sample Number 305-1 (μCi)	305-3 (μCi)	305-4 (μCi)	347-4 (μCi)
0.00 to 0.28[b]	1.55 ± 0.01 E-1	4.79 ± 0.03 E-1	Not measured	Not measured
0.28 to 8.23[c]	2 ± 1 E-4	1.31 ± 0.01 E+1	6.31 ± 0.05 E+1	1.48 ± 0.01 E+1
9.12 to 17.17	<4.0 E-5	4.9 ± 0.2 E-3	7.76 ± 0.07 E-1	1.95 ± 0.02 E+0
17.96 to 25.91	<3.2 E-5	2.0 ± 0.2 E-3	2.26 ± 0.06 E-2	1.06 ± 0.02 E-2

a. Activities as of 11/15/83.
b. This interval corresponds to the coatings layer.
c. This interval corresponds to the concrete just below the coatings layer.

In order to make a comparison between the results presented in Table 7 and the equivalent ^{137}Cs concentrations reported in Table 1, the paint and wafer activities were summed by sample and the total activities were converted to surface concentrations by dividing by the estimated cross sectional areas of the wafers. The areas used were 6.72 cm^2 for sections 305-1, 305-3, and 347-4 and 3.36 cm^2 for section 305-4. The calculated surface concentrations are shown in Table 8 under the heading "Table 7".

It is evident that the general agreement between the results obtained by the two different methods is not exact enough to allow a reliable calculation by subtraction of the activities in the coatings layers of sample sections 305-4 and 347-4. However, core 305-4 was among the six cores or core sections that were longitudinally gamma scanned.

To determine the resolution of the collimated spectrometer that was used to gamma scan the core samples a contaminated particulates filter was scanned while positioned at two different source-to-detector distances. Figures 10 and 11 show the count rate profiles that were obtained by gamma scanning the particulates filter. During the measurements, the plane of the filter was kept parallel to the plane of the collimator aperture so as to duplicate the geometry used during scanning of the core samples. In each figure, the count rate of the 662 keV photopeak of ^{137}Cs is plotted as a function of scan position. Figure 10 shows the results of the scan that was done while the filter was positioned with its near edge 2 cm away from the front surface of the collimator and Figure 11 shows similar results for the scan made using a counting distance of 3 cm.

As might be expected, both count rate profiles exhibit a sharp, symmetrical peak. A modest broadening of the peak with increasing counting distance is evident when the widths of the peaks are compared. The full width of the peak at one half its maximum value (FW 1/2M) increases from about 0.78 mm for counting done at 2 cm to about 0.94 mm for counting done at 3 cm. These graphs show the profile that should result from scanning a core sample whose sub-surface concrete is free of contamination.

The ^{137}Cs photopeak count rate profiles measured by gamma scanning whole core or core section samples 305-3, 305-4, 305-5, 305-8, 347-5, and

TABLE 8. COMPARISON OF ^{137}Cs SURFACE CONCENTRATIONS ($\mu Ci/cm^2$)

Sample	Table 1	Table 7
305-1	0.14	0.02
305-3	5.3	2.0
305-4	15.7[a]	19.0[b]
347-4	2.9	2.5[b]

a. Estimate based on 1000 mg/cm^2 TLD data.

b. Does not include contribution from paint.

347-8 are shown, respectively, in Figures 4 through 9. Sample 305-3 was a longitudinal half-section of the original core and sample 305-4 was a longitudinal three-quarter-section of the original core. The remaining four samples were intact whole cores.

Based on the assumption that the leading edge of each profile up to the maximum count rate corresponds to a measurement of activity in or very near the coatings layer, symmetrical peaks were drawn under the profiles of the four most contaminated samples (305-3, 305-4, 347-5, and 347-8) in order to estimate the partitioning of activity between the coatings layer and the sub-surface concrete. The FW 1/2Ms of the resulting four peaks are essentially identical and have an average value of 1.22 $\pm$ 0.04 mm. This width agrees reasonably well with the width predicted for a 4.3-cm diameter plane source.

The areas under the symmetrical peaks and under the entire profiles were determined and in each case the former was subtracted from the latter to estimate the fraction of the total activity residing in the concrete. Because the count rates plotted in the graphs were not corrected for sample self-absorption, the areas under the profiles corresponding to activity in the concrete were multiplied by correction factors and new total areas were computed. Approximate self-absorption correction factors were calculated using a linear attenuation coefficient at 662 keV of 0.184 cm^{-1} and, in each case, an attenuation thickness equal to one-half the core or section thickness.

The analysis described in the previous paragraph was performed on the count rate profiles of samples 305-3, 305-4, 347-5, and 347-8. The results, both uncorrected and corrected for self absorption, are presented in Table 9.

The percentages for sample 305-4 are presented as less-than and greater-than values because an insufficient length of the core was scanned to determine the total relative activity content of the concrete (see Figure 5). The relative activity content of the concrete of sample 305-4 determined by gamma scanning (20.4%) may be compared with the value that may be inferred using the results of the coatings removal decontamination

Table 9. ^{137}Cs PARTITIONING RESULTS

Sample Number	Activity Content (%)			
	Uncorrected		Corrected	
	Paint	Concrete	Paint	Concrete
305-3	91.2	8.8	89.6	10.4
305-4	<85.3	>14.7	<79.6	>20.4
347-5	98.0	2.0	97.0	3.0
347-8	63.2	36.8	53.6	46.4

experiment. Removing the paint from a quarter-section of 305-4 decreased its gamma exposure rate by 36%. Based on this result at least 64% of the total activity measured in the section is bound in the concrete.

The results presented in Table 9 for sample 305-3 indicate that about 10% of the ^{137}Cs activity is below the coatings layer on the half-section that was scanned. However, the results of radiochemical analyses performed on the paint and concrete wafers obtained from the mating half-section of sample 305-3 indicate about 95% of its ^{137}Cs activity is bound in subsurface concrete. The disagreement could be explained if the large majority of activity in this sample is distributed in a plane at or just below the interface of the coatings layer and concrete. The spatial resolution of the system used to scan the cores was too large to accurately resolve the partitioning in such a case.

Several of the count rate profiles show evidence of activity stratification within the core samples. This is particularly evident in the profiles of samples 305-3, 347-5, and 347-8. (See Figures 4, 8, and 9). Based on these profiles, the maximum depths that ^{137}Cs penetrated into the floor at these three locations are, respectively, 5.1, 2.3, and 2.8 mm. The count rate profile at core 305-4 indicates ^{137}Cs penetrated at this location to depths beyond 8.4 mm.

CONCLUSIONS

Results of analyses of surface samples collected prior to the Reactor Building gross decontamination experiment revealed that the fractions of the core inventories of fission products deposited on Reactor Building surfaces at the 305-ft elevation and above were less than 0.05%.[1] They also indicated that Cs and Sr were transported to Reactor Building surfaces in water droplets and that the majority of their measured surface activities were deposited after the nuclides had reached chemical equilibrium in the water in the basement.

The data obtained from this current study indicate that the protective coatings on concrete surfaces provided significant protection against radionuclide penetration. On the 305- and 347-ft elevation floors, radiocesium is generally confined within a few millimeters of the top surface of coatings layer. For this general case, the partitioning of radiocesium between the coatings layer and the subsurface concrete varies considerably. However, at floor locations having coatings that were damaged prior to the accident, radiocesium penetrated into the concrete to a depth of several centimeters. A breach or partial breach in the coatings layer combined with long-term pooling of contaminated water at the location of the damage is thought to be the cause of this phenomenon. The outward spread of sub-surface contamination at such locations appears to be confined to the near vicinity of the site of the damage.

Parametric calculations indicate that the decontamination of 347-ft elevation floor surfaces will significantly reduce the general area gamma exposure rates at this elevation. Measurements made of the depth of radionuclide penetration indicate that scabbling would be an effective decontamination technique.

The linear regression analysis results suggest that measurements of the contact beta and gamma exposure rates of a surface could be used instead of collecting samples from the surface to make a determination of the activity concentration on the surface. Because of the requirement that the survey instruments used for this purpose be highly collimated, the use of beta exposure rates is likely to be a more reliable method than one that uses gamma exposure rates.

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