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10 * Pages

JULY 5, 1946

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9-47

MEMORANDUM REPORT - DE-PC-164

SOURCE OF HIGH ACTIVITY LEVELS RECENTLY
OBSERVED FROM SHIPPING CONTAINERS AND TRANSFER CANS

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July 5, 1945

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I Introduction

Shipping cans containing product from plant runs completed in the latter half of April and in May showed, in general, definitely higher levels of radiation than had been observed previously. Likewise, containers for the transfer of the F-10-P solution from the 224 Bldg. to 231 Bldg. (during May) exhibited higher radiation levels than could be attributed to higher power levels and increased batch sizes.

These higher radiation levels were indicative that some radioactive element (or elements) was present in an amount greater than previously as would be expected from the shorter cooling periods, or that decontamination of elements previously present was less complete. It was considered essential to identify the source of these radiations and the cause for their presence has been investigated.

II Summary

1) The radioactive contaminant present in the final product has been identified rather definitely as Np^{239} which has a 2.34 half-life, a 0.28 Mev gamma ray and a weak beta ray. The amount of Np has increased as a result of the trend toward shorter cooling periods and the low decontamination factor for Np in the Separation Process.

The presence of Np does not present a serious health-hazard because of its rapid decay. The product of the decay is Pu^{239} .

2) The increase in the radiation levels from the cans for the transfer of the F-10-P solution from the 224 Bldg. to the 231 Bldg. observed in the latter half of April was due primarily to the increase in power levels and batch size.

3) In May, radiation levels as high as 160mr/hr (Run B-5-05-B-10) were observed from the transfer cans. This additional increase resulted from both higher power levels and the trend to shorter cooling times of 30-40 days instead of 40-50 days.

Roughly, two-thirds of the gamma activity (measured under standard counting conditions) in the P-1 solution of run B-5-05-B-10 was attributable to neptunium and most of the remaining third to lanthanum. Because of the relatively low energy of the Np^{239} gamma the major portion of the increased activity measured outside the transfer can was probably due to La.

Because of the rapid decay of each of these radioactive isotopes and because of the efficient removal of La in the isolation process the hazard is not considered to be serious.

III Radiation from Shipping Containers - Data and Discussion

Evidence was presented in SE-PC-76, File #3-1546, 3-8-45, and SE-PC-#18, File #3-2209, 4-20-45, which indicated that the product shipped from the Isolation Building (231 Bldg.) up until April 1, 1945 was essentially completely decontaminated. Radiation intensity at the surface of these containers which contained 80g of product amounted to 1-2mr/hr. This radiation was attributable to the gamma activity of product, itself.

The radiation from cans shipped in April and May has been as high as 30mr/hr and has averaged about 8mr/hr. A small part of this increase was due to the fact that the contents of the can were doubled (to 160g Pu).

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Experimental procedures to identify the contaminating radiation consisted of (1) estimations of the half-life and energy of the radiation from direct readings with electroscopes, (2) investigation of the radiations from AT solutions (final product solution from isolation process) by measurement of decay and absorption curves.

A. Electroscopes Readings

1. Decay of Radiation from Shipping Container

The Health Instrument Division was requested to monitor a sample can at intervals for as long a time as possible. Readings of Sample Can No. 88 containing 160g of product of which 17% was from run B-5-05-D-6 and 83% from run B-5-05-D-7 were taken over a three day period. Four readings were made at each interval, with the electroscopes placed at each of four quadrants of the can. The averages of these readings corrected for background are given in Table I.

Table I

Decay of Radiation from Sample Can 88

<u>Date</u>	<u>Time</u>	<u>Radiation Intensity in mr/hr at surface (aver- age of four readings at 90° angles)</u>
5-18-45	11:15 PM	10.1
5-19-45	7:00 AM	9.45
5-19-45	2:10 PM	8.9
5-19-45	11:45 PM	8.2
5-20-45	6:45 AM	8.5
5-20-45	2:20 PM	7.9
5-20-45	7:00 PM	7.75
5-21-45	7:00 AM	7.3

From these data the half-life of the radiation after correction for the radiation due to product is found to be about 80 hours.

2. Energy of Radiation from Shipping Container

A rough estimate of the energy of the radiation from the sample can was obtained by comparing Lauritsen electroscope measurements with and without 5g/cm² of Pb interposed between the electroscope and the sample can. For a representative sample can composed of 33% Pu from Run B-5-05-B-11 and 67% Pu from Run B-5-05-B-12, the intensity of 53mr/hr was reduced to 7.6mr/hr when the Pb was interposed. This corresponds to a reduction of gamma radiation intensity due to elements other than product by a factor of 4.4. In determining this factor it is necessary to subtract the background due to product. It was calculated that five g/cm² of Pb would reduce the intensity of 0.25 Mev gamma radiation by a factor of about 32, of 0.6 Mev, by a factor of 3.2, and of 0.8 Mev, by about 1.6. Therefore, it would appear that the energy of the radiation is less than 0.4 Mev. A more definite conclusion than this cannot be drawn since the geometry of the electroscope and lead absorber were such that some of the radiations did not pass through the lead before reaching the electroscope chamber.

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3. Radiation from AT Solutions (Final Product Solution)

The characteristics of the radiation from AT solutions due to elements other than product were determined using two methods of approach. 1) The level of beta activity was compared with the activity level due to the product and the decay of this activity was measured. 2) Samples of residue, left after the solvent extraction of product from 0.1 ml portions of AT solutions, were obtained from the Analytical Division as soon as possible after sampling. Since practically all of the fissile product activity should appear in the residue along with relatively small amounts of product, the activity background due to product was reduced to a minor level. Therefore, the accuracy of decay and absorption measurements was increased.

1. Measurement of Samples from AT Solutions

Samples were obtained from the AT solutions of runs T-5-05-B-4, 5, and 7, in order to determine whether or not the radiation levels from the cans in which these solutions were subsequently contained agreed with the relative levels of β and γ activity. From the table given below, it is evident that the magnitude of the increase in beta and gamma activity due to elements other than product is inadequate to account for the high electroscopes readings of 8 and 10 mr/hr. However, the samples were not counted until several days after the time when the electroscopes readings were made. Therefore, any short-lived activity would have decayed to an extent in accord with the activity found. This would confirm the indications mentioned above that the activity is short-lived. Samples were taken from May runs as soon as possible to permit measurement (before extensive decay) of the short-lived activities (see below).

Table II

Run No.	Electroscope Reading for Sample Can mr/hr	Activity in AT Solutions	
		Total beta activity Beta activity due to product	Total gamma activity Gamma activity due to product
Average Previous to April	1-2	1.00	1.0
T-5-05-B-4	8	1.14	1.5
T-5-05-B-5	10	1.01	1.2
T-5-05-B-7	10	1.07	1.5

2. Characteristics of Radiations from Residues from Solvent Extraction of AT Solutions.

The Analytical Division makes routine solvent extractions of product from samples of AT solution in order to determine gravimetrically the amount of fissile material present. Thus, solvent extraction residues were obtained from AT solutions of runs T-5-05-B-4 and 5, T-5-05-B-6, B-5-05-B-5, 6, and 7. The residues were dissolved in concentrated H_2SO_4 , transferred to Pt disks from which the activity was fixed, and then mounted for counting. In Table III are given the total β and γ activities in c/m/ml found in the residue from extraction of all samples, the β and γ activities due to the product in c/m/ml of AT solutions, and the ratios of the β and γ activity in the residue and that due to product alone.

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Table III

Activity Found in Solvent-Extraction Residues from AT Solutions

Run No.	γ c/m/ml (Residue)	γ c/m/ml Due to Product	Total γ Activity γ Activity Due to Prod.	β c/m/ml (Residue)	β c/m/ml Due to Product	Total β Activity β Activity due to Prod.
T-5-05-B-4	15	300	1.05	7.7×10^3	2.4×10^6	1.00
T-5-05-B-5	0	265	1.00	6×10^2	2.2×10^6	1.00
T-5-05-D-6	0	--	1.00			
B-5-05-D-5	17	203	1.09	1.4×10^4	1.7×10^6	1.01
B-5-05-D-8	0	164	1.00	5.2×10^5	1.3×10^6	1.00
B-5-05-D-7	0	--	1.00			

From these data it was apparent that the beta and gamma activities in these residues, which should contain most of the fission products present in the AT solutions were of negligible amount in comparison with the corresponding radiations of the product contained in the AT solution and insufficient in amount to account for the high electroscopes readings. Therefore, it seemed probable that most of the contaminating activity followed product in the solvent extraction.

Examination of the beta activity in the solvent extraction residues by Al-absorption measurements and decay measurements gave values of 2-4 days for the half-life and $\sim 13 \text{ mg/cm}^2$ for the half-thickness in aluminum.

C Conclusion

The identity of the radioactive contaminant present in the final shipping container is rather definitely established to be Np^{239} (element 93) which has a half-life of 2.3d, a 0.25 Mev gamma ray, and a beta ray with an initial $\sim 12 \text{ mg/cm}^2$ half-value thickness. Neptunium is known to follow plutonium in solvent extractions. The small amount of beta activity found in the residues results from the fact that the extraction is not 100% efficient. The properties of this isotope satisfy all the facts found in the studies described above. In addition, the amount of 93 associated with product prior to the Separations Process would increase as the cooling period is reduced. The decontamination of 93 in the Separations Process is known to be very low (a decontamination factor of around 200). Therefore, the amount of 93 associated with the final product would be much greater in runs made after April, in which the cooling period was reduced to less than 40 days, than in runs prior to the April series when cooling periods of 40-60 days were used.

The presence of element 93 in the final product is not injurious since it decays so rapidly that after a short time the radiation level from the sample can be essentially down to the background, due to product.

IV Radiation from Transfer Cans: 224 Bldg. to 231 Bldg.: Data and Discussion

Radiation levels from the cans in which the F-10-P solutions are transferred from the 224 Bldg. to the 231 Bldg. also increased during April. Readings as high as 40mr/hr were obtained in April ending high as 160mr/hr in May, as compared with readings of 5-10mr/hr prior to the April runs.

Some, but not all of this gradual increase in radiation level resulted from increased power levels and batch sizes. As in the case of the activity from the sample cans, the probable cause for the remainder of the increase in radiation level was the greater amounts of short-lived elements resulting from shorter cooling periods. Although the radioactive elements associated with product after the concentration process were essentially completely removed in the subsequent isolation process with the exception of element 93, it was considered important to determine whether the shorter cooling period or poorer decontamination was the cause of the higher level.

A Characteristics of Radiations from P-1 (P-10-P) Solutions

Activity in samples of P-1 solutions from runs T-5-04-B-4, T-5-04-B-6, and X-5-05-B-10 was examined by means of measurement of the decay and absorption curves to identify the predominant contaminant. The radiation from the transfer cans for these runs amounted to 3, 34, and 160mr/hr respectively. In the case of run X-5-05-B-10 an exceptional case was represented in that only 40 days elapsed between time of pushing from the pile and completion of the concentration process. The sample from this run was examined quite thoroughly.

Runs T-5-04-B-4 and B-6

Decay of Radiation: The curves obtained for the decay of the gamma radiation from the P-1 solutions of runs T-5-04-B-4 and B-6 were very similar to those described in SE-PC-118, File 13-2209, 4/20/45. As in the cases described in the reference, a short-lived component (half-life of about 3 days) constituted greater than 50% of the gamma radiation and mixtures of longer lived components (Sr, Cs, Ba, and product), the remainder. Likewise, the half-life of the contaminating beta radiation was about 14d in agreement with previous findings.

Half-thickness of Beta Radiation: Determination of the half-thickness of the beta radiation by absorption through Al. gave values of 38mg/cm² and 40mg/cm², respectively. This, together with the 14d half-life, established the fact that praseodymium was the predominant beta emitting contaminant, as in all previous runs examined.

Conclusions: The identity and relative amounts of the radioactive contaminants present were the same as in previous plant runs. The trend toward higher radiation levels from the transfer cans which started with run T-5-04-B-5 can be attributed primarily to increased batch-size and higher power levels.

Run X-5-05-B-10

The cause for the high radiation from the transfer can in this run (160mr/hr) appeared to lie in the short cooling period. If so, the short-lived elements, La and 93, might be expected to be present in proportionately greater amounts. To determine the identity of the contamination the following procedures were used.

- 1) The decay of the beta and gamma activity was followed.
- 2) Absorption curves were measured through Al and Pb to determine the approximate energies of the beta and gamma rays.

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Results: The decay curve and Al-absorption curve (Figure 1) for the beta radiation showed that more than 90% of the beta radiation had a half-life of less than 4d. About 75% had a half-thickness of 15mg/cm² of Al. Therefore, it is probable that almost 75% of the beta activity was due to element 93 (2.3d half-life, 12mg/cm² half-thickness).

The total gamma activity of the solution, immediately after sampling, was about 75 times greater than that due to product alone. This value is about three times higher than any observed previously.

The lead absorption curve for the sample is shown in Figure 2. Since the curve is complex, gamma radiations other than neptunium must be present. Decay measurements taken through 14.5g/cm² Pb showed an apparent 4 day half-life, indicating that a large part of the activity measured through this thickness of lead, was due to a short-lived gamma emitter, such as "orphan" lanthanum. On this basis it was possible to position an absorption curve, approximating that for lanthanum, so that the complex curve could be resolved. After making the graphical subtractions, a residual curve of ~1.5 g/cm² Pb half-value thickness was left. This value agrees with the half-value thickness normally observed for the 2.3 day neptunium activity, within the experimental error.

B. Conclusions

From the experimental data above, one may conclude that in the P-1 solution of run E-U-05-B-10, about two thirds of the gamma activity measured under standard counting conditions was due to ²³⁸Np. Most of the remaining activity was due to ¹⁴⁰La. However, for the conditions under which the transfer cans were measured in the 284 Building, the majority of the activity was due to the hard gamma radiation from the lanthanum. The amounts of both of these elements were high as a result of the short cooling times. Because of the rapid decay of each and the efficient removal of lanthanum in the isolation process, the hazard is not serious.

The experimental data for this report are recorded in Notebook E.K.W.-166-P.

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Al-Absorption Curve for Activity From P-1 Solution

3-2501

of Run 2-8-4-8-10

Figure 1

cpm/Sec

Added Absorber in mg/cm² of Al

10.

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15-Minute Decay for 100% Free I-131 Solution

of Run 3-8-55-1-10

Figure 2

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 1000 mV 1.5 A

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