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Z PLANT RADIATION STUDY SPECIAL SAMPLING PROGRAM INTERIM REPORT #2

by

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Z PLANT RADIATION STUDY
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Five kilograms of plutonium were removed from the Z Plant process stream during the week of February 10 - 14. Radiation measurements were taken on the process hoods prior to and during the actual sampling. Samples of nitrate, oxalate, oxide, fluoride, and metal, each containing from two to ten grams of plutonium, were sent to the Hanford Laboratory Operation Analytical Laboratory in 300 Area for analytical and spectrographic study.

Similar samples containing a minimum of 200 grams of plutonium were retained in Z Plant for measurement of variations in dosage rates with time and for evaluation of selected shielding materials.

The diverted plutonium being used in this special sampling program is currently scheduled for return to the process stream on March 31, 1958.

Gamma Energies

At the present stage of the study, gamma energies fall into two categories: (1) energies up to 0.45 mev and (2) energies above 0.45 mev.

Energies Below 0.45 Mev

The gamma energies of 0.45 and below represent the radiation from plutonium isotopes and the many decay products such as U^{237} and Am^{241} which result in the observed buildup of radiation in plutonium after purification. The study to date indicates that thorium 234 (UX_1) is another decay product which should not, as yet, be excluded as a radiation contributor in the 0.1 mev range.

Energies Above 0.45 Mev

Gamma energies above 0.45 which have been detected to date include the following:

1. 0.5 and 0.6 mev detected in the nitrate samples and attributed to ruthenium.

2. 0.75 mev detected in the nitrate samples and attributed to zirconium.

3. An energy of 1.27 detected in the fluoride sample and attributable to sodium 22, which is formed from fluorine through the alpha - n reaction. It emits a 1.28 mev gamma in decaying to neon 22.

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Since any appreciable gamma of these energies requires the thickness of lead glass for transparent shielding to be doubled in the Z Plant, it may develop that there is an economic justification for insuring a more consistently satisfactory removal of ruthenium and zirconium.

A more effective and consistent removal of the gamma contributors with energy of 0.5 mev and above would be aided by a specific index of these energies as a routine specification of material received from the separations plants. A modification to existing gamma scintillation counters to permit electronic cut-out of gamma energy below 0.45 mev is in the process of development by the Research and Engineering Operation. Data provided by this means to permit improved evaluation of fission product control may prove to be a valuable addition to the specification for Z Plant feed. Even without a tighter specification, it would appear to be of value as a continuing routine phase of the present Z Plant radiation study.

Process Hood Radiation

One of the recognized factors in control of radiation exposure is the quantity of radioactive materials in process. One phase of the study included a determination of the relative contribution of material in process to the total radiation from the hood. Both gamma and neutron radiation were determined at pre-established representative locations on the process hoods with the process vessels and equipment empty except for "heels" which could not be removed by "running the equipment dry."

Subsequent readings were taken on the hoods when sampled batch P-2-16 was in process. The readings from Hood 9B are presented in Table I.

TABLE I

Hood 9B Radiation Measurements

Ref. Point	Process Equipment Empty			Batch 2-16 in Process			Radiation Change by Process Material		
	Gamma	Neutron	Total	Gamma	Neutron	Total	Gamma	Neutron	Total
	mrem/hr			mrem/hr			mrem/hr		
4	14	8.6	22.6	16	12.0	28	+2	+3.4	+5.4
5	28	10.0	38.0	26	16.2	42.2	-2	+6.2	+4.2
6	20	7.3	27.3	17	8.8	25.8	-3	+1.5	-1.5
7	10	10.6	20.6	10	14.5	24.5	0	+3.9	+3.9
8	10	11.9	21.9	28	21.4	49.4	+18	+10.5	+28.5
9	10	13.7	23.7	13	25.5	38.5	+3	+11.8	+14.8
10	15	15.3	30.3	13	25.7	38.7	-2	+10.4	+8.4
12	14	15.4	29.4	12	21.4	33.4	-2	+6.0	+4.0

Radiation Contributed by In-Process Material

From data in Table I the following observations may be made for Hood 9B:

1. Emptying of the process equipment results in no overall reduction in gamma radiation.

2. Emptying of process equipment can reduce the neutron radiation by as much as 50 percent for Hood 9B.

Radiation Contributed by Hood Residue

There was no opportunity for noting the effect of cleaning hood interior surfaces during the sampling period of the study. However, data is available from earlier surveys on the effect of extensive hood cleaning on radiation levels.

Following a major cleaning of Hood 9B in November 1957, a conscientious effort was made to remove all radioactive material from the hood floors and interior equipment surfaces. Surveys taken on November 18, 1957, which reflected the result of this cleaning showed neutron radiations of 16, 18, and 22 mrem/hr and gamma radiations of 10 to 12 mr/hr with the process equipment full.

Comparing these readings with data in Table I (Batch 2-16 in process) there is little evidence of reduced radiation due to this extensive cleaning. The extreme difficulty of reaching the surfaces adequately to permit routine and efficient housekeeping of the hood interiors minimizes the potential of further reducing radiation from hood residues.

Neutron Shielding

In conjunction with gamma shielding tests, a measure of the attenuation of neutrons by iron was made using a PuF_4 sample as a neutron source. Although a reduction in energy was obtained, the equivalent mrem/hr of neutron radiation was not reduced by two inches of steel plate. The readings were 8.3 mrem/hr with energies of 0.92 mev and 0.75 mev respectively before and after attenuation through the two inches of steel.

Comparison of PuF_4 and $PuCl_3$ Neutron Radiation

A sample of plutonium chloride which has been prepared by the 234-5 Development Operation was compared for radiation readings with other plutonium compounds presently being studied. A table of comparative data is presented below.

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Pu Compound	Date Measured	Grams Pu	Neutron Radiation: Total mrem/hr at 6 in.	Neutron Radiation: E Mev	Neutron Radiation: mrem/hr 100 gr Pu
Chloride	3/21/58	421	0.55	0.97	0.13
Fluoride	2/18/58	328	33.4	1.0	10.18
Fluoride	3/3/58	328	24.8	0.92	7.56
Oxide	2/18/58	297	0.64	1.5	0.21
Oxide	3/3/58	297	0.45	1.5	0.15
Oxalate	2/18/58	299	0.93	1.7	0.31
Oxalate	2/28/58	299	0.69	1.7	0.23

The plutonium chloride ($PuCl_3$) emitted only 1.3 percent as much neutron radiation as the plutonium fluoride (PuF_4).

An apparent reduction in the neutron radiations with time will require further examination to insure against instrument variance or other extraneous factors. The higher neutron energies obtained for the oxalate and oxide as compared with the fluoride or chloride also require further investigation.

Conclusions

Conclusions from the study to date include the following:

1. The gamma radiation energies observed above 0.45 mev can be attributed to ruthenium and zirconium which are carried over from the separations plants and to the products of the alpha-n reaction of PuF_4.

2. Lead glass shielding must be twice as thick to provide the protection against the 0.75 mev gamma radiation of zirconium as is needed for radiation of 0.50 mev or below. Establishing methods for insuring removal of ruthenium and zirconium from the Z Plant feed would simplify the requirements of gamma shielding in at least part of the 234-5 process.

3. The chloride process, as now being developed, can reduce the neutron radiation now associated with the plutonium fluoride to less than 2 percent of the present exposure.

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4. Radiation outside of the hood from the process material moving through the equipment is small in comparison to that from the residual deposits throughout the hood interior.

5. The best efforts to date in reducing the hood radiation by cleanup of the hood interiors have been relatively ineffectual.

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