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Lung-Clearance Classification of ^{241}Am and $^{238-240}Pu$ in Dust Collected Near the Sites of Hanford Incidents 2827 and 2828

D. R. Kalkwarf
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Prepared for the U.S. Department of Energy
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SUMMARY

Dust samples containing ^{241}Am and $^{238-240}$Pu were obtained from sites near the occurrences of Hanford Incidents 2827 and 2828, and the alpha- activity ratios of ^{241}Am/$^{239-240}$Pu were found to be 215 and 1.7, respectively. The latter sample was also found to have a ^{238}Pu/$^{239-240}$Pu alpha-activity ratio of 0.033. The dissolution rates of these radionuclides into simulated lung fluid at 37°C were measured over a period of 10 days, and these measurements were used to classify the samples in terms of the lung clearance model proposed by the International Commission on Radiological Protection. The results showed that less than 0.02% of the $^{239-240}$Pu in both samples and less than 0.02% of the ^{238}Pu in the sample from Incident 2828 had dissolved in 10 days. Only 0.01% of the ^{241}Am in the sample from Incident 2827 and 0.27% of the ^{241}Am in the sample from Incident 2828 dissolved during this time period. It is recommended that the ^{241}Am and $^{239-240}$Pu components in both of these samples be considered 100% Y-type compounds for calculations with the ICRP Lung Clearance Model.

CONTENTS

INTRODUCTION

The purpose of this study was to classify the lung clearance of ^{241}Am and $^{239-240}Pu$ from dust samples collected near the sites of Hanford Incidents 2827 and 2828 in terms of the ICRP Task Group Lung Model. The International Commission on Radiological Protection developed this model for use in computing the radiation dose from radionuclides deposited in the lung. A key parameter is the solubility classification of the deposited material defined according to the rate at which it leaves the lung. Three classes were established: D, W, and Y, corresponding to half-times in the lung of 0 to 10 days, 11 to 100 days, and >100 days, respectively. If clearance of the material from the lung is not strictly exponential with time, it is approximated by a sum of exponentials; and the material is classified according to the fractions of D, W, and Y components. In the absence of biological data, lung-clearance half-times for materials have been approximated by their dissolution half-times in simulated lung fluids.[1-4] Although endocytosis and ciliary-mucus transport are known to contribute to lung clearance, experiments have indicated that a few days after dust deposition, dissolution determines the clearance rate for the lower respiratory tract.[5,6] Given the lung-clearance classification for a material, its transport rates between other anatomical compartments are automatically assigned in the model. From these parameters, one can compute the residence times of the material and the associated radiation dose in each compartment.[7]

In the present study, the dust samples were obtained from Radiation Monitoring personnel who collected the samples after the incidents had occurred. The sample obtained from the site of Incident 2827 was collected immediately after the incident had occurred whereas that obtained from the site of Incident 2828 was collected several days later, when the area had been cleared for entrance.

Dissolution-rate classifications were based on measurements of the fraction of actinide remaining undissolved in a sample as a function of time in simulated lung fluid at 37°C. Maximum dissolution rates were sought by means of rapid agitation of the samples because the lung is expected to be a site for efficient dissolution and because the values were to approximate clearance rates that include contributions from endocytosis and ciliary-mucus transport.

CONCLUSIONS AND RECOMMENDATIONS

The dust samples from Hanford Incidents 2827 and 2828 were found to have $^{241}Am/^{239-240}Pu$ alpha-activity ratios of 215 and 1.17, respectively. The latter sample was also found to have a $^{238}Pu/^{239-240}Pu$ alpha-activity ratio of 0.033. The dissolution results showed that less than 0.02% of the $^{239-240}Pu$ in both samples had dissolved in 10 days. Similarly, less than 0.02% of the ^{238}Pu in the sample from Incident 2828 dissolved within 10 days. Only 0.01% of the ^{241}Am in the sample from Incident 2827 and 0.27% of the ^{241}Am in the sample from Incident 2828 had dissolved during this time period. It is recommended that the ^{241}Am and $^{239-240}Pu$ components in both of these samples be considered 100% Y-type compounds for calculations with the ICRP Lung Clearance Model.

PROCEDURE

PREPARATION OF SAMPLES

The sample obtained from the site of Incident 2827 was received as dust particles contained in a plastic Petri dish whose cover had been sealed to the dish along its periphery with plastic tape. Due to the small amount of material, no attempt was made to subdivide the sample according to particle size. The dish was opened while enclosed in a large plastic bag, located inside a hood; and the dish was inverted over a plastic funnel so that the dust sample fell through it into a 5-ml, conical bottom vial (Pierce Chemical Co., Reacti-vial) with a V-shaped Teflon stirrer. After this transfer, 5.00 ml of freshly prepared simulated lung fluid at pH 7.3 and 37°C were added to the vial, it was capped with a Teflon-lined, screw cap, and placed in a constant-temperature block/stirrer (Pierce Chemical Co., Reacti-Therm) at 37°C.

The sample obtained from the site of Incident 2828 was received as dust particles imbedded in a 4x4-inch asbestos-fiber filter. The surface of the filter was vacuumed with a plastic filter holder (Millipore Corp., Swinnex-25) loaded with a 25-mm diameter membrane filter with 0.01 μm pores (Millipore Corp., VF) and attached to a vacuum line. Based on measurements of the filters alpha activity with a hand-held survey meter (Eberline, Model 21039) before and after this operation, it was estimated that approximately 35% of the activity was removed. This dust was transferred to a 5-ml conical-bottom vial with a

V-shaped Teflon stirrer, and 5.00 ml of freshly prepared simulated lung fluid at pH7.3 and 37°C were added. The vial was then capped and placed in the same constant-temperature block stirrer as described above.

PREPARATION OF SIMULATED LUNG FLUID

The electrolyte compositions of human interstitial lung fluid and the simulant used in this study are shown in Table 1. Comparison shows that they are almost identical. The protein components of actual lung fluid were represented by an ionically equivalent amount of citrate in the stimulant as suggested by Moss.[8] Lung-fluid proteins are poorly characterized and generally not available in large quantities, and substitute proteins hinder filtration and promote bacterial growth in solutions. Phospholipids, also known to be present in trace amounts in actual lung fluid, were not included in the simulant for the same reasons.

TABLE 1. Compositions of Actual and Simulated Lung Fluids

Ion	Actual[8]	Simulated[9]
Calcium, Ca^{2+}	5.0 meq/ℓ	5.0 meq/ℓ
Magnesium, Mg^{2+}	2.0 "	2.0 "
Potassium, K^{+}	4.0 "	4.0 "
Sodium, Na^{+}	145.0 "	145.0 "
Total Cations	156.0 "	156.0 "
Bicarbonate, HCO_3^{-}	31.0 meq/	31.0 meq/
Chloride, Cl^{-}	114.0 "	114.0 "
Citrate, $H_5C_6O_7^{3-}$	--	1.0 "
Acetate, $H_3C_2O_2^{-}$	7.0 "	7.0 "
Phosphate, HPO_4^{2-}	2.0 "	2.0 "
Sulfate, SO_4^{2-}	1.0 "	1.0 "
Protein	1.0 "	--- "
Total Anions	156.0 "	156.0 "
pH	7.3-7.4	7.3-7.4

Simulated lung fluid with the composition shown in Table 1 was prepared by slowly adding the following ingredients in order to 990 ml of distilled water and adjusting the final volume to 1000 ml:

0.2033 g $MgCl_2 \cdot 6H_2O$
6.0193 g NaCl
0.2982 g KCl
0.2680 g $Na_2HPO_4 \cdot 7H_2O$
0.0710 g Na_2SO_4
0.3676 g $CaCl_2 \cdot 2H_2O$
0.9526 g $NaH_3C_2O_2 \cdot 3H_2O$
2.6043 g $NaHCO_3$
0.0970 g $Na_3H_3C_6O_7 \cdot 2H_2O$

If the pH of the resulting solution was not 7.3-7.4, it was adjusted to this value with small volumes of 1 N HCl.

DISSOLUTION TECHNIQUE

Dissolution trials on the dust samples were conducted in well-agitated portions of simulated lung fluid (SLF) at 37°C. Dissolution started when SLF was added to the samples in their container vials. After selected time periods, each vial was removed from the heating block and centrifuged to force the undissolved dust into the conical end. The cap was then opened, and the supernatant fluid was drawn through a stainless steel needle into a plastic syringe. A membrane filter (Millipore, 13-mm diameter, GC, 0.22 μm pore size) in a stainless steel filter holder (Millipore, Swinnex) was fitted on the end of the syringe, and the solution was filtered into a container and stored for the actinide analyses. The membrane filter was then removed with stainless steel forceps, and 5.00 ml of fresh SLF was added to the barrel of the syringe. The filter holder, minus filter, and the syringe needle were refitted on the syringe; and the small amount of solid sample held on the filter was washed off into the vial with a jet of SLF from the syringe. The vial was then capped, vortexed to resuspend all the remaining sample and replaced in the heating block. The pH of the suspension was checked every three days and adjusted to 7.3-7.4 with dilute HCl, if necessary. At the end of 10 days, the residual sample was dissolved in 5.00 ml of warm concentrated nitric acid and analyzed for ^{238}Pu, $^{239-240}Pu$ and ^{241}Am.

ANALYSES FOR ^{241}Am AND $^{239-240}Pu$

^{241}Am was determined by direct gamma-ray spectroscopy. Standard solutions of ^{241}Am in nitric acid were pipeted into 2-dram screw-capped vials and their volumes were adjusted to those of the samples. These standards and the dissolution samples were then counted for 59.5 KeV gamma rays on a Ge(Li) diode. The activities in the samples were calculated from their counting rates and the disintegration/count factor determined from the standards.

^{238}Pu and $^{239-240}Pu$ were determined by alpha energy analysis following chemical separation. The separation procedure consisted of dissolving plutonium in nitric acid and purifying it by anion-exchange chromatography.[10] Radiochemical yields were determined by tracer techniques using ^{242}Pu.

RESULTS

DISSOLUTION BEHAVIOR OF SAMPLES

The results of the dissolution trials are listed in Tables 1 and 2. They show that less than 0.02% of the $^{239-240}Pu$ in both samples dissolved within 10 days. Similarily, less than 0.02% of the ^{238}Pu in the sample from Incident 2828 dissolved within 10 days. Only 0.01% of the ^{241}Am in the sample from Incident 2827 and 0.27% of the ^{241}Am in the sample from Incident 2828 dissolved during this time period. By evaluating the total amounts of ^{241}Am and $^{239-240}Pu$ in each sample, the $^{241}Am/^{239-240}Pu$ ratios were found to be 215 for that from Incident 2827 and 1.17 for that from Incident 2828.

TABLE 1. Incremental Amounts of Radionuclide Dissolved and Fraction, F, of Designated Radionuclide Remaining Undissolved During Dissolution of the Sample from Hanford Incident 2827

Time	^{241}Am	$^{239-240}Pu$	F_{Am}	F_{Pu}
0.00 days	-	-	1.0000	1.0000
3.00	$(1.72\pm.02)\times10^{4}$ d/m	$483\pm$ 2d/m	1.0000	.9998
7.00	$(6.68\pm.12)\times10^{3}$	lost	1.0000	.9998
10.00	$(2.59\pm.02)\times10^{4}$	$7.7\pm.2$	.9999	.9998
Undissolved	$(5.30\pm.01)\times10^{8}$	$(2.47\pm.02)\times10^{6}$		
Total	$(5.30\pm.01)\times10^{8}$	$(2.47\pm.02)\times10^{6}$		

TABLE 2. Incremental Amounts of Radionuclide Dissolved and Fraction, F, of Designated Radionuclide Remaining Undissolved During Dissolution of the Sample from Hanford Incident 2828.

Time	^{241}Am	^{238}Pu	$^{239-240}Pu$	F_{Am}	F_{Pu}
0.00 days	-	-	-	1.0000	1.0000
4.00	62±17d/m	<0.05d/m	0.14±.07d/m	.9993	1.0000
7.00	150±21	<0.02	0.17±.05	.9976	1.0000
11.00	26±15	0.14±.02	5.4±.1	.9973	.9999
Undissolved	$(8.84\pm.03)\times10^4$	$(2.53\pm.01)\times10^3$	$(7.57\pm.08)\times10^4$		
Total	$(8.84\pm.03)\times10^4$	$(2.53\pm.01)\times10^3$	$(7.57\pm.08)\times10^4$		

CLEARANCE CLASSIFICATION OF SAMPLES

Clearance classifications were based on the dissolution behavior of the samples, and dissolution half-times were used as estimates of lung-clearance half-times. If a compound has this classification Y in the ICRP Lung Clearance model, a fraction greater than $F = \exp(-0.693t/100)$ should remain undissolved after t days of dissolution. Table 3 shows values of F for various values of t.

TABLE 3. Values of $F = \exp(-0.693t/100)$

t	F
1 day	.09931
3	.9794
4	.9727
7	.9526
10	.9330
11	.9266

Since all of the values for F_{Am} or F_{Pu} in Tables 1 and 2 exceeded these values at a given time, it was concluded that the ^{241}Am and ^{238}Pu and $^{239-240}Pu$ components in both samples are class Y compounds.

DISCUSSION

The chemical natures of the ^{241}Am, ^{238}Pu and $^{239-240}Pu$ in the dust samples were not known. Experiments with dogs have shown that $^{239}PuO_2$ moves very slowly from the lung,[11] and that a clearance classification of Y is to be expected for this compound. A sample of $^{241}AmO_2$, on the other hand, was found to exhibit a substantial amount of class W behavior in dogs.[12] The lack of class W behavior in either of the samples from Incidents 2827 and 2828 may be due to differences in the chemical or crystallographic form of the ^{241}Am or to differences in the particle size distribution, and hence specific surface areas, of the samples.

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