LUNG RETENTION AND TISSUE DISTRIBUTION OF INHALED ^{239}Pu AEROSOLS PRODUCED AT DIFFERENT TEMPERATURES

ABSTRACT

Lung retention and tissue distribution of inhaled ^{239}Pu aerosols produced at temperatures of 325, 600, 900 and 1150°C were studied in Beagle dogs for 56 days duration. Aerosol characterization showed no differences in activity median aerodynamic diameter, geometric standard deviation or in estimated density among the various treatment temperatures. Lung retention and tissue distribution were determined in 3 animals at each aerosol production temperature and found to be influenced by production temperature. At the lower two temperatures, significant translocation of ^{239}Pu to liver and skeleton occurred whereas at the higher temperatures, only very small quantities of ^{239}Pu were found in extrapulmonary tissues. Translocation of ^{239}Pu from lung to tracheobronchial lymph nodes was not affected by aerosol production temperatures.

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INTRODUCTION

Increasing quantities of ^{239}Pu will be handled as mixed-oxide fueled reactors and liquid metal fast breeder reactors are added to our nation's energy production facilities. The development of substantially increased fuel reprocessing, in which the ^{239}Pu will be radiochemically separated and reclaimed, may increase the potential for human inhalation exposure either occupationally or environmentally to aerosols to ^{239}Pu with different *in vivo* solubilities. The present study was designed to examine the effect of heat treatment temperature on the subsequent biological behavior of an inhaled ^{239}Pu aerosol. It was conducted concurrently with another study designed to study the effects of temperature treatment on the subsequent efficacy of various therapeutic measures in the event of an accidental exposure to a polydisperse ^{239}Pu aerosol (this report, pp. 269-273).

MATERIALS AND METHODS

The polydisperse aerosols used in this study were nebulized from a solution of $^{239}PuCl_4$ in 1 M HCl with a Pu concentration of 3.0 mg/ml. The solution was filtered immediately prior to usage each day. All aerosols were subjected to a two-stage heat treatment in which the first stage was maintained at 325°C and the second stage at 325, 600, 900 or 1150°C. respectively, for the 4 exposure groups. During each exposure, the aerosol was sampled by filter to determine mean activity concentration, by cascade impactor to determine particle aerodynamic size distribution; and on electron microscope grids using a point-to-plane electrostatic precipitator to determine the particle physical size and shape. An estimate of particle density was made by combining aerodynamic and physical size measurement. A sample of the aerosol produced at each temperature was subjected to *in vitro* solubility tests as described elsewhere in this report (pp. 269-273).

This study involved 12 Beagle dogs ranging in age from 29 to 34 months such that 3 dogs were exposed by inhalation at each aerosol treatment temperature in an apparatus previously described (1972-1973 Annual Report, LF-46, pp. 10-15). Equal numbers of both sex were used and the duration of the inhalation exposure ranged from 9 to 24 mintues. Animals were housed individually in metabolism cages after the inhalation exposure for collection of urine and feces throughout the experimental period. At 56 days post-inhalation exposure, they were sacrificed by exsanguination under pentobarbital anesthesia and tissue samples were obtained for radiochemical analyses. These tissues and

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all excreta samples were subjected to alternating dry and wet ashing analytical radiochemistry procedures to produce a clear aliquot for internal sample liquid scintillation counting to quantify ^{239}Pu.

Plutonium-239 does not emit sufficiently penetrating radiations for routine external measurement of lung or whole-body radioactivity. In this study, and its complement, the initial lung burden and its retention were estimated from daily excreta samples and tissue activity determined after sacrifice. Fecal excretion represents mechanical clearance of material by ciliary transport from the respiratory tract to the esophagus, swallowing and transport through the gastrointestinal tract. Urinary excretion represents the elimination of a fraction of material solubilized in the lung and transported in blood. The remaining fraction of material solubilized in lung and transported in blood is deposited in other tissues. To reconstruct the initial lung burden and its retention pattern, it was assumed that urine activity in each daily sample represented a constant fraction of the blood activity on that day. The value for this partition coefficient between urine and blood was estimated by taking the ratio of total urine activity collected from day 3 through day 56 to the total activity in tissues other than lung. Using this coefficient a value for solubilized activity leaving the lung was estimated for each day. Summation of the daily fecal, urinary and blood ^{239}Pu activities was employed to reconstruct the initial lung burden and lung burden retention.

RESULTS AND DISCUSSION

Aerosols produced at the four temperatures were not discernibly different by electron microscopy, all being near spherical in shape with a slight surface roughness. The measured activity median aerodynamic diameters (AMAD) and geometric standard deviations (σ_g) were not significantly different for the four temperature treatment aerosols, having a mean of 1.9 μm AMAD and 1.8 σ_g for all exposures. Aerosol density estimates varied rather widely about a mean of 4.8 gm/cm^3 but were not significantly different for the four temperature treatments. The *in vitro* solubility studies conducted on aerosol samples produced at the four temperatures showed a decreased solubility as temperature of production increased for the three eluants tested (this report, p. 269).

Reconstructed lung retention data are shown in Figure 1 for each of the 3-dog groups exposed at the four aerosol production temperatures. These data indicate that lung retention decreased as temperature of aerosol production decreased corrolating well with the observed *in vitro* solubility data. The predicted long-term retention of ^{239}Pu derived by fitting exponential functions to the data by a non-linear least squares method indicate effective retention half-lives of 1250, 750, 340 and 180 days, respectively, for aerosol production temperatures of 1150, 900, 600 and 325°C. These data indicate that greater differences exist in the solubility of the two lower temperature aerosols while the 900°C and 1150°C aerosol are nearly indistinguishable when such long half-lives are assessed in a 56-day period.

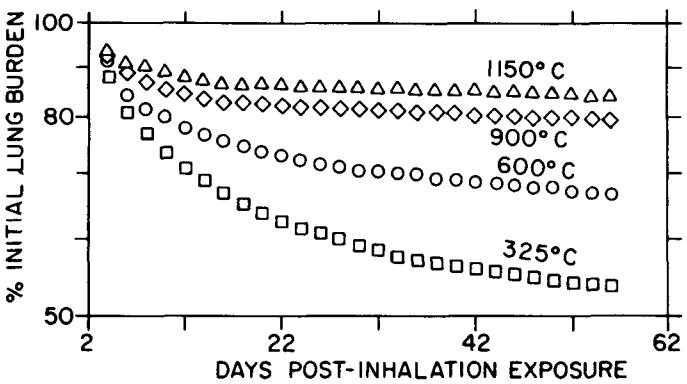

Figure 1. Retention of ^{239}Pu in dog lung expressed as a percent of the initial lung burden for aerosols produced by heat treatment of $^{239}PuCl_4$ at 4 different temperatures. Each set of data represents the mean of 3 values.

Table 1 presents the percent of sacrifice body burden found in selected tissues at 56 days post-inhalation exposure. The lowest aerosol production temperature yielded the highest translocation of ^{239}Pu from lung to liver and skeleton with decreasing quantities found in these tissues as aerosol production temperature increased. Significant in its deviation from this trend is the relative uniformity of ^{239}Pu content in the tracheobronchial lymph nodes at all aerosol production temperatures indicating transport processes to these regional lymph nodes are not related to aerosol solubility.

Table 1

Percent of Sacrifice Body Burden Found in Selected Tissues Following Inhalation of ^{239}Pu Produced at Four Temperatures

Aerosol Temp.	Percent of Sacrifice Body Burden				
	Lung	Liver	Skeleton	TBLN	Other
325°C	74	12.0	12.1	0.4	0.9
	86	4.9	7.7	1.4	0.4
	78	7.7	13.1	0.3	0.9
600°C	97	0.1	1.9	0.9	0.1
	94	2.1	2.9	1.0	0.3
	92	3.0	3.8	0.9	0.5
900°C	99	0.1	0.3	0.4	0.2
	99	0.1	0.3	0.1	0.1
	99	0.1	0.2	0.6	0.1
1150°C	99	ND[a]	0.6	0.5	0.1
	99	ND	0.1	1.3	0.3
	99	ND	0.1	ND	0.8

[a]ND = Non Detectable

This study has served to emphasize the importance of aerosol particle solubility in the metabolism of ^{239}Pu following inhalation exposure. As discussed elsewhere in this report (pp. 269-273), aerosol particle solubility also affects the efficacy of various therapeutic procedures. The extent to which aerosol particle solubility also influences the development of biological response to inhaled ^{239}Pu aerosol is receiving increased attention at this Institute.