• Toxicology of Plutonium-Sodium

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Scenarios for liquid-metal fast breeder reactor (LMFBR) accidents predict the loss of sodium coolant, with subsequent core melt-down and release of mixed sodium-fuel aerosols [Na-(PuU)O_2] into the environment. Studies in other laboratories demonstrated that mixed aerosols of Na_2O-PuO_2 were more readily transported from the lung than PuO_2 aerosols. We therefore devised a continuous aerosol-generating system for animal exposures in which laser-generated fuel aerosols were swept through sodium vapor to form sodium-fuel aerosols. These fuel and sodium-fuel aerosols were compared with regard to their physicochemical properties and their biological behavior following inhalation studies in rats and dogs.

Previous studies concerning physicochemical characterization of laser-generated fuel aerosols revealed that they were enriched in uranium and oxygen relative to the fuel. The fuel aerosols consisted of branched-chain aggregates of very small particles with an activity median aerodynamic diameter (AMAD) of <1 μm. Sodium-fuel aerosols were spherical, with AMADs proportional to the sodium-fuel ratio and larger than those of the fuel aerosols.

In vitro solubility of the mixed aerosols was greater than that of fuel aerosols. Enhanced solubility, in vivo, of the mixed aerosols was indicated by the appearance of larger fractions (compared to those of fuel aerosols) of the initial lung burden in blood, liver and skeleton of rats or dogs following inhalation exposure.

Chemical studies designed to explore mechanisms for the enhanced solubility of the fuel-sodium aerosols revealed that uranium was rapidly converted to a soluble uranyl carbonate complex-ion species in the presence of air and moisture. A small quantity of atomically dispersed plutonium results from destruction of the fluorite-type lattice when uranium dissolves. Results from investigations of plutonium oxidation states in sodium-PuO_2 aerosols are consistent with the chemical reactions $Na + 3PuO_2 \rightarrow Pu_2O_3 + NaPuO_3$.

It was apparent from chemical studies that release of a more soluble form of Pu into the environment could significantly alter deposition and distribution of Pu in pregnant individuals. A previous study (Annual Report, 1979) involving inhalation exposure of pregnant rats late in gestation (19 days) indicated that fetoplacental depositions of ^{239}Pu from sodium-fuel aerosols exceeded that of the fuel aerosols. We have continued these studies to determine fetal deposition patterns following maternal exposure to smaller sodium-fuel particles (2.4 versus 3.5 μm) at earlier stages of gestation.

In the current study, we exposed Wistar rats at 15 or 19 days gestation (dg) for 1 hr, nose-only, to a LMFBR-fuel aerosol or a sodium-fuel aerosol (Table 1). Groups of 5 or 6 rats were killed immediately after exposure, at 1 day, or at 5 days for ^{239}Pu analysis of maternal and fetal tissues. Results from these analyses were compared following normalization of aerosol values to 40 nCi/L.

TABLE 1. Characteristics of LMFBR-Fuel and Sodium-Fuel Aerosols.

Characteristic	Fuel	Sodium-Fuel
Sodium-Fuel Ratio	0:1	40:1
^{239}Pu Concentration, nCi/L	210	40
AMAD/GSD[a]	0.66/1.50	2.40/1.41

[a]Activity median aerodynamic/geometric standard deviation.

Initial lung burdens for maternal rats inhaling fuel aerosols appeared lower than

those of rats exposed to the sodium-fuel but were not significantly different (Table 2). Five days after exposure, lung values for rats exposed to sodium-fuel were significantly lower than initial lung burdens. By comparison, lung clearance of fuel aerosols was somewhat reduced. Maternal blood values for ^{239}Pu in sodium-fuel-exposed rats were markedly higher than those of the fuel-exposed rats during the entire experimental interval. These differences were reflected in the ^{239}Pu values for components of the fetoplacental units (FPU).

Plutonium deposition within the FPU was highest in membranes, next highest in placenta, and lowest in fetuses (Table 3). Plutonium-239 accumulated more readily in the larger, more-developed structures of the 19-dg than in the 15-dg FPU. Values for the 20-dg FPUs were not significantly different, although the 15-dg sodium-fuel-exposed FPU appeared to acquire more ^{239}Pu over the 5-day exposure than did the 19-dg FPU during a 1-day exposure. These results demonstrate that fetal accumulation of ^{239}Pu following exposure to the more-soluble sodium-fuel aerosols is 60 to 80 times greater than fetal deposition following fuel-aerosol exposure. The data also imply that exposure to these aerosols earlier in fetal development, or even prior to implantation, may enhance fetal accumulation of ^{239}Pu during the longer period of intrauterine exposure.

The results of this project clearly demonstrate that in order to fully evaluate the potential effects of Pu released to the environment from reactor accidents, it is important to understand the biological behavior of complex mixtures of the sodium actinides.

TABLE 2. Lung and Blood Values (Mean ± SE) in Pregnant Rats Following Exposure to Fuel or Sodium-Fuel Aerosols (Normalized to 40 nCi/L).

Tissue	Days After Exposure	Aerosol	
		Fuel	Sodium-Fuel
Lungs, nCi	0	45 ± 3[a]	62 ± 8[a]
	1	47 ± 4[a]	57 ± 6[a]
	5	38 ± 3[a,b]	29 ± 3[b]
Blood, pCi/ml	0	0.34 ± 0.18[a]	17.1 ± 2.9[c]
	1	0.15 ± 0.04[a]	5.8 ± 0.9[b]
	5	0.16 ± 0.06[a]	5.8 ± 0.8[b]

[a-c]Values with common letters for a given tissue are not significantly different ($P < 0.05$).

TABLE 3. Distribution of ^{239}Pu Within the Fetoplacental Unit of Rats that Inhaled LMFBR-Fuel or Sodium-Fuel Aerosols (pCi, Mean ± SD).

Tissue	Days After Exposure	Fuel[a]		Sodium-Fuel[a]	
		15 dg[b]	19 dg[b]	15 dg[b]	19 dg[b]
Placenta	0	0.04 ± 0.01[c,d]	0.01 ± 0.002[c]	2.2 ± 0.3[e]	6.1 ± 1.6[e,f]
	1	0.02 ± 0.003[c]	0.05 ± 0.005[d]	3.1 ± 0.4[e]	16 ± 2.5[f,g]
	5	0.14 ± 0.04[d]		17 ± 1.6[g]	
Membranes	0	0.02 ± 0.005[c]	0.02 ± 0.002[c]	0.5 ± 0.1[e]	3.6 ± 1.5[c,d,e]
	1	0.02 ± 0.003[c]	0.11 ± 0.02[d]	5.5 ± 0.9[f]	34 ± 4.7[g]
	5	0.46 ± 0.16[c,d,e]		60 ± 8.4[g]	
Fetus	0	0.05 ± 0.02[c,d]	0.04 ± 0.01[c,d]	0.3 ± 0.06[e,f]	0.7 ± 0.2[f]
	1	0.01 ± 0.002[c]	0.03 ± 0.006[c,d]	0.2 ± 0.02[e]	1.9 ± 1.9[g]
	5	0.03 ± 0.003[d]		2.5 ± 0.5[g]	

[a]Aerosol activities normalized to 40 nCi/L.
[b]Days of gestation at exposure
[c-g]Values with common letters for a given tissue are not significantly different ($P < 0.05$).