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Acute toxicity of uranium hexafluoride, uranyl fluoride and hydrogen fluoride

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ACUTE TOXICITY OF URANIUM HEXAFLUORIDE, URANYL FLUORIDE AND HYDROGEN FLUORIDE

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ABSTRACT

Uranium hexafluoride (UF_6) released into the atmosphere will react rapidly with moisture in the air to form the hydrolysis products uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF). Uranium compounds such as UF_6 and UO_2F_2 exhibit both chemical toxicity and radiological effects, while HF exhibits only chemical toxicity. This paper describes the development of a methodology for assessing the human health consequences of a known acute exposure to a mixture of UF_6, UO_2F_2, and HF.

1. INTRODUCTION

Uranium hexafluoride (UF_6) released into the atmosphere will react rapidly with moisture in the air to form the hydrolysis products uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF). The corrosive HF vapor formed by this reaction has a pungent odor and is very irritating to the skin and mucous membranes. The soluble uranium compounds, UO_2F_2 and UF_6, exhibit both chemical toxicity and radiological effects.

Individuals exposed to these toxic materials may suffer varying health effects depending upon the concentration of the toxicant, the duration of the exposure and many other factors. For example, an accident evaluation may require consideration of the ability of personnel to escape quickly, the variation in the spatial concentration of the toxicant(s), and the physical activity level at the time of exposure. Although these factors may be important when evaluating the hazard associated with an accidental UF_6 release, the information presented in this report does not attempt to account for all the many variables that may need to be considered in a hazard evaluation. Rather, this report focuses on predicting the health effects given the exposure duration and toxicant concentration.

2. CHEMICAL TOXICITY OF UF_6, HF and UO_2F_2

In 1980 a group of experts in the field of chemical toxicity of soluble uranium and HF were asked to apply known data and make their best judgments about the toxicological effects of postulated exposures to soluble uranium and HF. This information was then used to develop preliminary Design and Analysis guidelines for estimating the toxicity of soluble uranium and HF.

A review of the information obtained during the development of the preliminary Design and Analysis guidelines indicated a lack of directly applicable data for assessing the consequences of acute UF_6, UO_2F_2 and HF exposures. Therefore, it was concluded that it would be desirable to obtain additional data on the consequences of acute exposures to UF_6 and UF_6 hydrolysis products. The U. S. Department of Energy (DOE) sponsored a series of animal toxicity experiments at the University of Rochester in order to provide additional data to support accident assessments of U. S. UF_6 handling facilities.

The primary objective of the toxicity experiments was the development of a procedure for evaluating the consequences of acute exposures to mixtures of UF_6 and UF_6 hydrolysis products. This goal was achieved by completing the following tasks:

(1) determination of the lethal exposure susceptibility of rats and guinea pigs to UF_6 and the UF_6 hydrolysis products UO_2F_2 and HF;

(2) definition and measurement of delayed effects of uranium and fluorine in animal survivors of UF_6, UO_2F_2 and HF exposures; and

(3) prediction from the results of the animal experiments, minimum exposure levels for humans of UF_6 and UO_2F_2/HF mixtures which will result in significant physiological damage for short periods of exposure.

In late 1983 the experimental work was completed and documented in a report submitted by the University of Rochester to Martin Marietta Energy Systems, Inc.(1) As indicated in Table 1, a total of 66 experiments were conducted utilizing 511 rats and 78 guinea pigs for exposure durations ranging from 2 to 60 minutes. The results of the University of Rochester rat and guinea pig experiments are summarized in Table 2.

A "Delphi" panel of toxicologists was formed to interpret the experimental results. J. B. Hursh, L. J. Leach, and P. E. Morrow of the University of Rochester and M. E. Wrenn of the University of Utah were asked to develop independent, preliminary estimates of the toxicity of UF_6 hydrolysis products. These preliminary toxicity estimates were presented at a meeting where each

[1]Operated by Martin Marietta Energy Systems, Inc. for the U. S. DEPARTMENT OF ENERGY under Contract No. DE-AC05-84OR21400.

Table 1. Summary of University of Rochester Animal Experiments

No. and Species	No. of Experiments	Exposure Duration (min)	Air Concentration in g-U/m³ - Mortality Relationship			
			LC50[a]	95% Confidence Interval	LC10[b]	95% Confidence Interval
150 Rats	15	2	120.0	99.3 - 140	55.0	40.0 - 76.0
170 Rats	17	5	38.6	26.8 - 55.7	10.5	6.48 - 17.1
140 Rats	14	10	12.0	10.1 - 14.3	5.16	3.65 - 7.29
51 Rats	7	60	0.74	0.49 - 1.10	0.27	0.12 - 0.47
78 Guinea Pigs	13	2	62.1	43.4 - 88.8	13.5	5.45 - 33.5

(a) Concentration corresponding to 50% lethality.

(b) Concentration corresponding to 10% lethality.

Table 2. Dose-Response Relationships for Rats and Guinea Pigs

Species	50% Lethal Concentration (g-U/m³)	Exposure Duration (min)	Concentration-Time Product (g-U/m³)(min)
Rat	120	2	240*
Rat	38.6	5	193
Rat	12.0	10	120*
Rat	0.74	60	44
Guinea Pig	62.1	2	124

*These numbers are statistically different and indicate that in the rat studies the concentration-time product does not have a constant value for a given biologic response (percent mortality).

toxicologist presented his approach and rationale for estimating the toxicity associated with acute UF_6, UO_2F_2 and HF exposures. The toxicologists were then asked to reevaluate their toxicity estimates, if necessary as a result of the discussions, and to submit documentation describing the rationale used in developing their "final" estimates of toxicity. The final toxicity estimates were then used to develop a basis for assessing the toxicity of soluble uranium and HF.(2)

The basis derived by the panel of toxicologists for assessing the toxicity of soluble uranium compounds such as UF_6 and UO_2F_2 is presented in Table 3 and on Figure 1. Four health effect classifications were established for characterizing the toxicity of soluble uranium compounds: (1) no effect, (2) possible mild health effects, (3) renal injury, and (4) lethality.

The possible mild health effects regime corresponds to exposure levels that are greater than the no effect level and less than the renal injury level. An exposure predicted to result in possible mild health effects may result in observable biological effects, but such exposures will not, in themselves, result in either a short term or a long term impairment in the body's ability to function. The renal injury classification indicates that significant physiological damage to the kidneys is predicted. The lethality health effect classification corresponds to an exposure that is expected to result in 50% mortality. Estimates of uranium toxicity for exposure times greater than 60 minutes should be based on extrapolation of the 60-min toxicity estimates as shown on Figure 1.

Table 3. Chemical Toxicity of Soluble Uranium Compounds

Health Effect	Uranium Absorption[a] (mg-U/kg)	Exposure Level[b] (mg-U/m³)(min)	Exposure Time (min)
50% Lethality	1.63	35,000	Indefinite
Renal Injury	0.058	1,250	≤ 30 min
		750	≥ 60 min
No Effect	0.03	650	≤ 30 min
		390	≥ 60 min

[a]The absorbed quantity of uranium per kg of body weight.

[b]The exposure level is defined as the product of the airborne and the exposure time. Based on the ICRP resting respiration rate of 7.5 L/min.

Estimates of HF exposure levels as related to various physiological effects are presented in Table 4 and on Figure 2. Five health effect classifications have been established for exposures to HF: (1) no effect, (2) smell/no health effects, (3) smell/possible irritation, (4) irritation/possible health effects, and (5) lethal. An HF exposure is predicted to have no effect if the HF concentration is less than the detection by smell level.

3. RADIOTOXICITY OF URANIUM

Uranium-234, uranium-235, and uranium-238 are alpha emitters of widely varying specific activity. U-234 has a specific activity approximately 2900 time that of U-235 and 18,000 times that of U-238. Thus, U-234 has the greatest radiotoxic potential. For example, at 3% U-235 enrichment, the U-234 represents only 0.222 wt.% of the total uranium present, but it is responsible for more than 78% of the total activity and, hence, total radiation dose.

Based on experiments in which small animals were subjected to plutonium exposure, it appears that the minimum health effect resulting from acute exposure to alpha radiation is a decreased immune response, which could occur when the total activity in the lung exceeds 100 μCi.(3)

Table 5 depicts the relative chemical toxicity and radiotoxicity of uranium that has been enriched to 97.5% U-235 and 1.14% U-234. It is evident, from the data shown in Table 5, that for acute exposures to soluble uranium, radiotoxicity is negligible in comparison with chemical toxicity, even at 97.5% U-235 enrichment and 1.14% U-234 enrichment. Therefore, it is concluded that the radiotoxicity of an acute uranium exposure is insignificant when compared with the associated chemical toxicity.

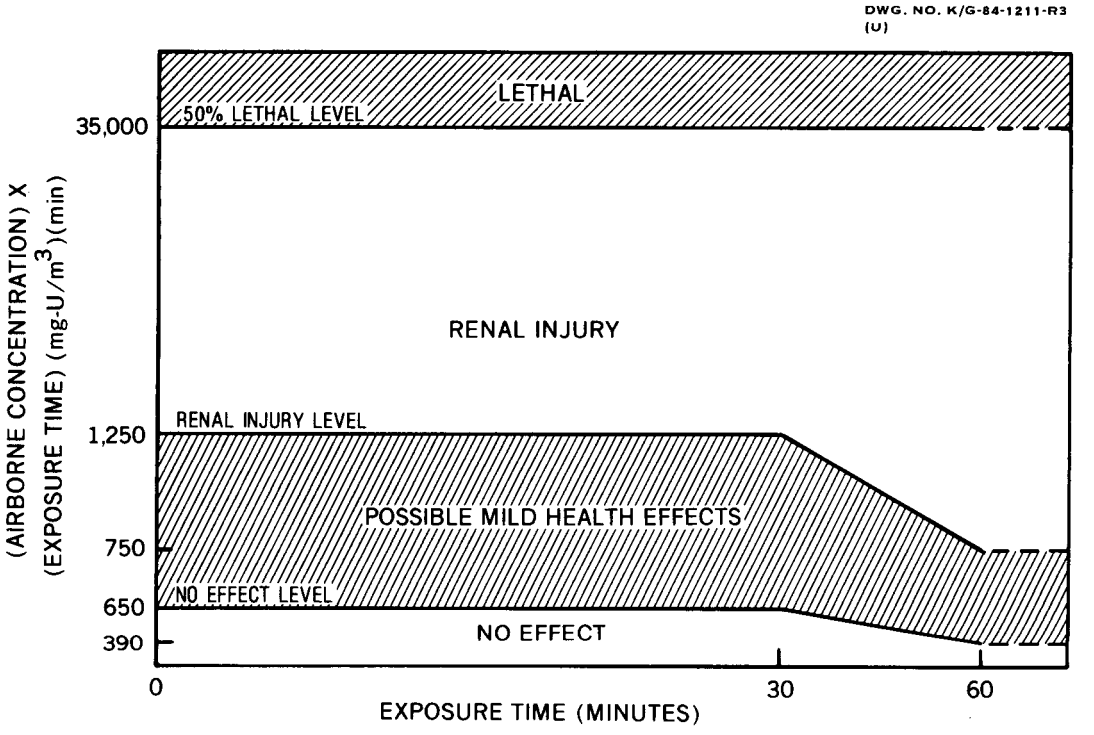

Figure 1. Toxicity of Acute Exposures to Soluble Uranium.

Table 4. Toxicity of Hydrogen Fluoride

Health Effect	HF Concentration (mg/m^3)	Exposure Time
Detection by Smell	1	Indefinite
Smell/No Health Effects	2.5	$\leq$ 8 hr
Irritation	26	< 10 min
	13.33	$\geq$ 10 min
Lethal*	$\frac{53{,}000}{\text{exposure time (min)}}$	0 to 60 min

*Estimates of HF lethality are based on an inhaled exposure of 53,000 $(mg\text{-}HF/m^3)(min)$.

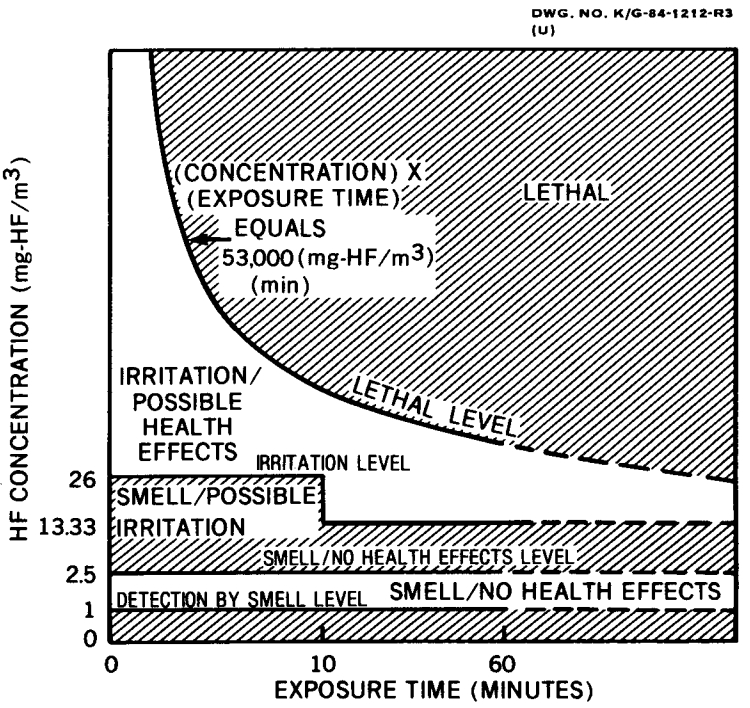

Figure 2. Toxicity of Acute Exposures to Hydrogen Fluoride.

Table 5. Comparison of Chemical Toxicity and Radiotoxicity of Soluble Uranium*

Absorbed Dose of Soluble Uranium (mg-U/kg)	Equivalent Radiation Dose (μCi)	Acute Health Effects	
		Chemical Toxicity	Radiotoxicity
0.03	0.16	No effect	No effect
0.058	0.30	Renal injury	No effect
1.63	8.45	50% lethality	No effect
19.29	100.	Lethal	Onset of radiological effects

*At 97.5% U-235 and 1.14% U-234 enrichment.(4)

4. REFERENCES

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4. Personal communication between A. C. Bassett (Goodyear Atomic Corporation) and R. A. Just, August 1984.