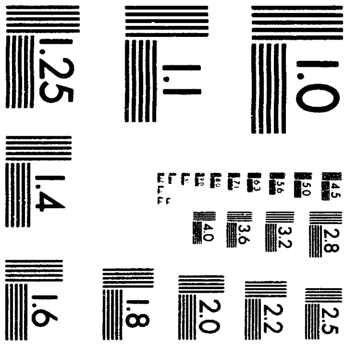
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AIRSOURC Code Aerosol Model in Radioactive Waste Tanks

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AIRSOURC CODE AEROSOL MODEL IN RADIOACTIVE WASTE TANKS

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ABSTRACT

An aerosol model was developed for a radioactive waste tank vapor space and ventilation system. The source of aerosol formation was modeled from gas bubble bursts at the liquid surface. The gases contained in these bubbles are formed by radiolytic breakdown of liquid and organic materials in the radioactive tank contents. The model accounts for the rate of radiolytic gas formation and the rate of tank liquids that are released as aerosols. The concentration of particles in the tank vapor are modeled as a function of the deposition rate of dry particles and the vapor removal rate by ventilation. The ventilation rate in tanks without an active ventilation system has a threshold level that is driven by atmospheric pressure variation.

The model predictions are compared to measurements in several typical waste tank vapors of the Hanford Site waste storage tanks.

INTRODUCTION

The processing wastes from chemical separation of irradiated reactor fuel are stored in large underground tanks on the Hanford Site. These tanks contain radioactive fission products from the fuel and chemicals, such as nitrates, used in the chemical separation process. A source term model of volatile vapors and aerosol emissions from these tanks was developed to provide information for environmental licensing and equipment upgrade decisions. This model was developed to determine emissions from specific tank conditions and, thus, supplement nonsystematic methods specified by the licensing agencies. Modeling the aerosols and their releases from the tanks will be the subject of this paper.

DISCUSSION

The double-shell waste storage tanks used for storage of liquid irradiated reactor fuel processing waste are on the order of 1-million-gallon capacity. The older single-shell tanks are smaller. Filtered single-pass ventilation systems are installed on the double-shell tanks. Some of the single-shell tanks have filtered vents that are not actively ventilated. The model of aerosol release source terms considered radionuclide concentrations in the liquid. These aerosols arise from operational activities, such as tank filling, air sparging, and liquid transfer, and are affected by differences in ventilation rates. In addition, an aerosol source is also iniated by small gas bubbles formed by radiolytic breakdown of water and other chemical compounds in the tank waste. The following descriptions are given of these models.

Splashing Aerosol Model

This model estimates the fraction of tank liquids made airborne by splashing from an above-surface fill nozzle. The correlations used are described in Ballinger et al. (1988). They are based upon correlating experimental spill data with the Archimedes Number. The fraction of aerosol generated is given by:

$$\text{Aerosol Fraction} = 8.9\times10^{-10} \times ARCH^{0.55} \qquad (1)$$

where

ARCH = Archimedes Number.

The Archimedes Number was derived through dimensional analysis of liquid and slurry spill parameters (Ballinger et al. 1988) and is defined as:

$$ARCH = \frac{\rho_{air}^2 \times H^3 \times g}{\mu_l} \qquad (2)$$

where

ρ_{air} = density of air (g/cm^3)
H = distance from leak to splash surface (cm)
g = gravitational constant (980.7 cm/s^2)
μ_l = liquid viscosity, poise (g/cm·s).

In situations where the leak is being discharged with a finite downward velocity (rather than free fall), the spill height must be adjusted as follows:

$$H' = H + \frac{V^2}{2 \times g} \qquad (3)$$

where

V = downward leak velocity (cm/s),

which is determined from the flow rate and discharge area of the fill nozzle.

Sparging Air Aerosol

This model calculates the aerosol generated by radiolytic gas formation and the air sparging systems installed in some of the tanks. This model was developed from an effervescence model, used for other applications, that determines the aerosol from acid reaction with concrete walls. The aerosol formation from air sparging and radiolytic gas formation in the tank is equal to the aerosols formed by effervescence. The air sparging and radiolytic gas model will use the air equivalent volume to the carbon dioxide (CO_2) produced by acid effervescence.

In the effervescence model, the aerosol is generated from a spill of aqueous acid solution. The aerosol is generated when the acid contacts a concrete surface and reacts, causing the spill solution to effervesce when CO_2 gas is created.

Data in Ballinger et al. (1987) indicate that ≃ 5% of the spill volume is released as aerosol per mole of CO_2 released. Therefore, if it is assumed that all liquids have an aqueous base with 55.5 g/mol total solution per liter, then the volume fraction of the spill released as aerosol is

$$Aerosol\ Fraction = \frac{0.05 \times CO_2}{V \times 55.5} \tag{4}$$

where
V = liquid volume (ℓ), and
CO_2 = quantity of CO_2 gas (mole).

This equation can be related to the mass of aerosol produced by air sparging by equating the volume of 1 mole of CO_2 to 22.4 ℓ (0.0224 m^3) air at standard temperature pressure.

$$R = (Aerosol\ Fraction) \times V \tag{5}$$

$$= \frac{0.05 \times 0.0224 \times V}{V \times 55.5} \tag{6}$$

$$= 2.02 \times 10^{-5}\ m^3. \tag{7}$$

Passive Ventilation Rate

In tanks where an active ventilation system is not available, the atmospheric pressure variations will cause an exchange rate of outside air with the tank vapors. A representative annual exchange rate is 1.69 atmospheres per year (Crippen 1993). The passive air exchange rate of a 100,000 ft^3 vapor space will be:

$$F_p = \frac{1.69\ vol/yr \times 100{,}000\ ft^3/vol}{365 \times 24 \times 60\ \text{min}/yr} \tag{8}$$

$$= 0.32\ ft^3/\text{min}. \tag{9}$$

Release of Aerosols with Deposition Correction

For a given particle size, the deposition rate can be described as an exponential

$$c = c_o e^{-\lambda t} \tag{10}$$

where
c = time-dependent concentration
c_o = initial concentration
λ = removal coefficient
t = time (h).

For a steady state situation,

$$V\frac{dC}{dt} = R - \lambda CV - F_oC = 0 \qquad (11)$$

where

R = release rate aerosol formation rate
F_o = ventilation flow out
V = vapor volume.

The average concentration will be:

$$C = \frac{R}{\lambda V + F_o}. \qquad (12)$$

The release rate from the tank vapor to the exhaust system is:

$$F_oC = \frac{F_oR}{\lambda V + F_o}. \qquad (13)$$

Deposition Removal Coefficient

Aerosols from two sources were considered. One source was from fill liquid splashing. The other source was from bubbles formed by air sparging and radiolytic breakdown of tank liquids. The mean wet droplet size of splashing and air sparging were assumed to be 28.8 and 3.0 microns, respectively. The droplets will quickly evaporate leaving a dry particle of smaller diameter, which will have a smaller rate of deposition. The volume fraction of dissolved solids was assumed to be no greater than 4% in the aerosols. A particle that is reduced to 4% of its original volume by evaporation will have a dry diameter that is 34% of is wet diameter. The corresponding dry particle diameters of the splashing and sparging or bubble-formation aerosols are 9.8 and 1.0 microns, respectively.

The tank vapor and geometry was modeled by the FPFP_2 code (Owczarski 1991) for these two mean particle diameters. The model was based on a five-particle diameter distribution about the mean diameter. The deposition rates of these two mean particle depositions, represented by Formula 10, give values of the removal coefficient corresponding to:

λ = $-0.39\ h^{-1}$ for sparging bubbles
= $-15.6\ h^{-1}$ for splashing aerosols.

Radiolytic Gas Formation Rate

Radiolytic gas is formed by ionizing radiation absorbed in the tank materials. An assumption was made that the gas conversion factor, expressed as a yield "G" factor, is 0.2 molecules/100 eV radiation absorbed. Because of the large volume of the waste tanks, almost all of the energy is absorbed by the tank waste. The reduction in dose rate near the tank edges by escaping gamma radiation was ignored. The radiolytic gas volume formation rate was added to Formula 4 above.

* Results and Conclusions

A calculation of aerosols was made for a single-shell waste tank. Some of the tank parameters are listed in Table 1.

The concentrations in the tank vapor volumes were determined for several significant radioisotopes. Examples of these results are given in Table 2.

These results were generally conservative and were in reasonable agreement with preliminary data.

Table 1. Calculational Parameters of a Waste Tank.

Temperature	22.8 °C
Liquid volume	250 m^3
Vapor volume	2,700 m^3
Liquid transfer rate	0 gpm
Liquid surface	410 m^2
Air sparging rate	0
Radiolytic gas formation	6.8 E-3 m^3/h
Air ventilation rate	0.54 m^3/h

Table 2. Aerosol Concentrations in Waste Tank Vapor.

Isotope	Liquid concentration (Bq/m^3)	Vapor concentration (Bq/m^3)
^{90}Sr	3.0 E+12	3.9 E+2
^{137}Cs	6.0 E+8	7.9 E-2
^{239}Pu	1.8 E+10	2.3 E+0
^{241}Am	6.0 E+9	7.9 E-1

The modeling techniques discussed in this paper predict tank emissions systematically from individual tank conditions and operational parameters. From an engineering standpoint, this approach is more satisfactory than the nonsystematic regulatory method. This model will provide guidance for operational and equipment upgrade considerations.

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