MASTER

THE RESTRICTED RELEASE OF PLUTONIUM

by

D. C. Hunt

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
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GOLDEN, COLORADO 80401
U.S. ATOMIC ENERGY COMMISSION
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Erratum in
 "The Restricted Release of Plutonium"

A units conversion factor error occurs in equation (8, p.17. The factor g_c in equation (8 was assumed to be 980 whereas in the c.g.s. system of units used in the paper, it should be unity. The free release source strength values obtained in the paper may be qualitatively scaled in a conservative manual as follows:

- A. **Pressure Dependent Calculations** - The conservative assumption is made that the "fixed" value of TCON [equation (22;p.23)] is increased by a factor of 22.5. This implies a "fixed" explosive dispersion model in which all of the energy involved in the plutonium oxidation heats the enclosure air. The air flow from the enclosure will then be turbulent if $\lesssim 15$ kg of plutonium is oxidized and will be fully turbulent for oxidation of the "fixed" mass of 202 kg. Under these conditions the source strength magnitude is principally controlled by the factor $V_d L / V_i d$ specified by equations (17 and (18 on p.28. The V_i for fully turbulent flow is given by equations (8, (9-c, (22, (23, and (24. The quantity $V_d L / V_i d$ will then be increased by a factor of 2.36 due to an increase of a factor of 1.45 in the flow velocity, a decrease of a factor of four in the friction factor and an increase of a factor of about 22.5 in the leak path diameter. This will lead to a decrease in the turbulent non-deposition probability by a factor of about 4×10^4 . The number of leak paths now required to normalize the calculations to the 0.5 V/o per hour leakage rate is increased by a factor of about 31 while

the leak path entrance area is increased by a factor of 500. The net change in the safety factor of 15.4 is an increase of a factor of 2.65 so that the scaled safety factor is now 40.8.

- B. Pressure Independent Calculations - The scaling in these calculations is straightforward since the leak path dimensions are assumed independent of the enclosure overpressure. Assuming explosive dispersion of the plutonium in which only about 2.5% of the enclosure air acts as a heat sink (i.e. assuming a TCON scaled up a factor of 980 from the "fixed" value) the source strength variation results shown in Figure 11-a. are still valid.

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ABSTRACT

A study is made of the possible hazard outside an enclosure due to the uncontrolled oxidation of plutonium within the enclosure. The report first reviews the existent observational data on such "restricted" plutonium release and then constructs a release model relating the "free release" source strength and the parameters describing the release. The method of analysis is to postulate a set of accidents in which the release variables are individually maximized. The accident which the model indicates as having the greatest release potential is taken as the maximum credible accident. The method is used to evaluate the leakage limits of the critical mass assembly cell of the Rocky Flats Plant Nuclear Safety Facility. The results generally indicate that restricted release is unlikely to lead to dangerous air-borne plutonium concentrations and specifically show the Rocky Flats Plant Assembly Cell leakage limits are conservative by several orders of magnitude.

THE RESTRICTED RELEASE OF PLUTONIUM

A. INTRODUCTION AND SUMMARY

This report studies the "restricted" release of plutonium, that is, release from within an enclosure in which plutonium is handled. The purpose of the study is to determine the relation between the leakage rate of air from the enclosure and the resultant air-borne concentrations of plutonium outside the enclosure under several sets of release conditions. The questions of any economic loss due to the release or the mechanism of plutonium uptake and deposition in the body are not considered. Thus no plutonium deposition on surfaces outside the enclosure is assumed and the 40 hour/week continuous exposure air-borne concentration limits in AEC Manual Chapter 0524 are accepted. These limits for Pu^{239} are shown in Table I. The concentration units are $\mu\text{ci/ml}$:

TABLE I

PLUTONIUM AIRBORNE CONCENTRATION LIMITS		
	<u>Controlled Area</u>	<u>Uncontrolled Area</u>
Insoluble	4×10^{-11}	1×10^{-12}
Soluble	2×10^{-12}	6×10^{-14}

The report first reviews some pertinent observational data on restricted plutonium release and then constructs a model of restricted release. The model is able to predict "free release" source strengths as a function of the following variables:

1. The assumed leak path models.
2. The particle size distribution.
3. The mass of plutonium involved in the release.
4. The surface-to-mass ratio of the plutonium involved in the release.
5. The release rate of plutonium.

6. The mode of particle transport to the enclosure walls
7. The type of thermal equilibrium achieved in the enclosure during the release.

The free release source strengths are combined with existent atmospheric dispersion theories (e.g. Gifford, 1961) to predict the maximum air-borne concentration possible for a given source strength.

Sets of accidents are then postulated in which the release variables are individually maximized with the other variables maintained at very severe "fixed" values. The accident which, according to the model, has the greatest associated air-borne plutonium concentration is assumed to be the maximum credible accident. This maximum credible source strength is compared with the appropriate Table I limit. * The model is normalized so that the computed air-borne concentrations are consistent with whatever enclosure leakage rates are being evaluated.

The above procedure is somewhat subjective but it is shown as the model is developed that it is almost certainly conservative, i.e., tends to overestimate the resultant air-borne plutonium concentrations for a given leak rate, so that the model should be satisfactory for hazards calculations. The model is applied to the specific case of the critical mass assembly cell of the Rocky Flats Plant Nuclear Safety Facility.

* See Appendix III

B. OBSERVATIONAL DATA ON RESTRICTED PLUTONIUM RELEASE

The data available on restricted plutonium release is of two types; first, observations are available on plutonium release associated with the accident experience of several plutonium handling facilities, and second, several experiments have been performed on the release, or the simulated release, of plutonium.

1. Observations of Accidental Release

a. Oxidation of Metallic Plutonium

Tuck and Hunt (1965) have discussed the observations of plutonium release associated with accidents at the Rocky Flats Plant of The Dow Chemical Company. In one incident (September, 1957) approximately 22 kg of plutonium spontaneously ignited in a glove box. The glove box walls burned and the fire also destroyed all filters between the box and the building exhaust stack while the exhaust fans were running at high speed in an attempt to remove smoke from the room. Even though the entire mass of plutonium was oxidized, environmental air and vegetation sampling showed increases in activity which were well below hazardous levels.

In another Rocky Flats Plant incident an 800 g plutonium briquette spontaneously ignited while it was being weighed in a glove box. A dry box glove was removed to allow attempts to extinguish the fire but the briquette burned completely. The continuous air effluent sampler near the box indicated an average plutonium concentration of 2×10^{-13} mci/cm³ for the 2 hour period during which the incident occurred. No measurable amounts of plutonium were found at environmental air sample stations nor from soil or water samples.

b. Explosive Dispersion of Plutonium in Solution

On November, 1959, a chemical explosion occurred at ORNL in an evaporator containing plutonium within a shielded

cell (King and McCarley, 1961). About 150 g of plutonium (probably the entire evaporator contents) was dispersed to the cell and about 0.6 g was blown out the cell door. Buildings and nearby streets were contaminated and the operating area of the building was contaminated by the air flow through open pipes and other cell wall penetrations. About 1.5 g of plutonium was removed in the cell ventilation filter plenum and no measurable amount of plutonium was released via this system to the plant stack.

c. Dispersion of Plutonium From Burning Metal in a Hydrocarbon and Organic Solvent Environment

On October 15, 1965, a fire occurred at Rocky Flats Plant in a lathe dry box drain line containing oily plutonium chips and turnings. The fire involved 20 to 30 g of plutonium and occurred when sparking resulted from an attempt to clear the clogged oil drain line. The spread of plutonium from the fire caused air-borne concentrations to exceed the 8 hour radioactivity concentration guide throughout almost all of Buildings 76 and 77. The prevailing spread was along established building ventilation air flow patterns and as far as 190 feet from the fire the air activity was greater than 100% of the 8 hour concentration limit. No measurable activity occurred beyond the buildings even though some penetration of the absolute filters in the main building filter plenum occurred. The analysis of the filter paper from the air sample heads (located between 15 feet and 50 feet from the fire) indicated the air-borne particles were mostly PuO_2 even though oils and an organic solvent recently used to clean the drain line (CCl_4) were present at the site of the fire. The great spread of the particles is probably due largely to the fine state of division of the metal involved in the release but may also be associated with the catalyzing action of the oil and the CCl_4 .

The apparent greater dispersion of plutonium associated with the solution and hydrocarbon incidents appear to indicate that smaller particles, which are more easily transported, are involved than in the metallic burning cases. These particles are probably oxides but could also be hydrides, carbides, nitrides, or compounds such as chlorides associated with process solvents. In the Rocky Flats lathe box fire, for instance, the measured particle mass median diameter was 0.32μ , or a factor of about 10^3 smaller in diameter than measured PuO_2 particles formed from burning plutonium (Stewart, 1963).

2. Experiments on Restricted Release

a. Gravel Gertie Experiments

In order to measure contamination due to explosions involving fissile material, the AEC designed a special test structure known as Gravel Gertie. The distinctive feature of Gravel Gertie is a 15 foot gravel covering, supported by steel cables and steel mesh, which is used instead of a solid roof. In the event of a detonation within the structure involving fissile material it was expected that the roof would raise, absorbing some of the explosive shock and filtering the nuclear material from the escaping gas. A schematic of the structure is shown in Figure 1. The primary interest of the Gravel Gertie tests was to determine if containment could be achieved in an explosion involving plutonium. However the difficulties and expense involved in a plutonium shot precluded its use, so that uranium was finally substituted as a tracer material. This substitution was justified by the data obtained in a prior shot conducted in the open involving both uranium and plutonium in which the deposition data for the two elements compared quite favorably (Cowen, 1964). In the final Gravel Gertie test, conducted in April, 1957, the entrance to the structure was sealed and a charge of uranium was detonated with

550 pounds of high explosive. The gravel roof of the structure was observed to raise a maximum amount of 22 feet and did not settle back into place for about 3 seconds. As a result of previous explosions in the structure, many cracks and missile craters existed in the walls, and large cracks were found in the two 10 foot cubical bays (see Figure 1). Also the ceiling cables failed under the dynamic load of the falling rock, filling the room with gravel. The results of this test were that no measurable amount of uranium was dispersed outside of Gravel Gertie even under these rather extreme release conditions.

b. Exploding Wire Experiments

Cheever (1961), performed a set of experiments which involved volatilizing 1 or 2 g amounts of plutonium in the form of a slab cylinder or small turning. The ignition was done in a plutonium system glove box which was exhausted at a rate of 5 cfm through a series of 3 to 7 high efficiency filters. The results of the tests were that an average of 0.00055% of the plutonium burned was collected on the filters and that an average penetration of the first filter of 0.5% occurred. The average particle size of the plutonium oxide aerosol was found by electron microscope studies to be 0.016μ .

The conclusions to be drawn from the experiments and accident experience involving plutonium release is that "restricted" release of plutonium is not likely to be large. Even in the case of the ORNL accident, when 150 g of a probably very finely divided aerosol of plutonium was dispersed explosively into a cell, only 0.6 g escaped through the open cell door and the spread of plutonium contamination outside the cell was not excessive.

C. THEORY OF RESTRICTED RELEASE

1. Probable Type of Release

The released plutonium is assumed to consist entirely of Pu^{239} as this is the most hazardous of the plutonium isotopes in the fuel (cf. AEC Manual Chapter 0524). The most likely compound present in an accidental plutonium release is plutonium dioxide (PuO_2). This is due to the plentiful supply of oxygen which would probably be associated with a substantial plutonium release and to the relatively large reaction heat for dioxide formation (252 K cal/mole) which energetically favors PuO_2 as a reaction product. Large releases of other plutonium compounds would be likely only under special release circumstances such as a chemical explosion in a system where plutonium metal is immersed in a non-oxygen containing organic liquid. Also any volatilization of plutonium or PuO_2 will not contribute significantly to the air-borne hazard (Hilliard, 1961).

With regard to hazards calculations, it is not extremely important what plutonium compounds are involved in a release. Any dependence of free release source strength on the compound type may be accounted for by the ranges of particle size distributions and release rates which are assumed. Thus for the sake of definity all releases are assumed to consist entirely of PuO_2 particles of density 11.45 g/cm³.

It should be noted here that even though the Gravel Gertie experiments were quoted to emphasize the small probability of restricted release under extreme release conditions, the model does not assume any initial impulse overpressure associated with the release. This is because the purpose of the report is not to assess the ability of an enclosure to withstand an explosion but to evaluate a certain enclosure leakage rate specified at a certain overpressure. Thus the plutonium release itself must not be allowed to change the enclosure leakage rate other than through a change in the static overpressure associated with the release.

2. Particle Size and Mass Distributions

Several determinations of size and mass distributions of PuO_2 particles released from plutonium metal have been made (Stewart, 1963; Anderson, 1964; Kirchner, 1966). These distributions vary widely depending on the crystalline phase of the metal, the oxidation temperature, the humidity, temperature, motion and composition of the ambient gas, the surface to mass ratio of the metal, and (unfortunately) on the method of measurement used. Any special oxidation circumstances such as oxidation from solution will also strongly affect particle distributions. In order to easily use measurements of particle size and mass, it is customary to assign the particles an "equivalent spherical diameter." Such an assignment then uniquely specifies the relation between the mass and size distributions. Depending on the method of measurement, this assignment will give a particle distribution which somewhat overemphasizes inertial processes and underestimates diffusive transport and thermophoresis or vice versa. Thus for any reasonably polydisperse particle distribution the use of the equivalent diameter concept should not lead to serious error. For theoretical calculations it is very convenient to have an analytical expression for the particle distributions. Many aerosols, including PuO_2 particles under some release circumstances (Anderson, 1964), exhibit a so called log normal distribution (Fuchs, 1964) or:

$$g(\delta)d\delta = \frac{0.399}{\ln\sigma_g} \frac{1}{\bar{m}} \frac{4}{3} \pi \rho \left[\frac{\delta}{2} \right]^3 \exp \left[-\frac{1}{2} \left\{ \frac{\ln(\delta/\bar{d}_m)}{\ln\sigma_g} + 3 \ln\sigma_g \right\}^2 \right] d \ln \delta \quad (1)$$

where $g(\delta)d\delta$ is the fraction of the total particle mass associated with particles whose equivalent spherical diameter is in the range δ to $\delta + d\delta$, $\bar{d}_m$ is the mass median diameter, $\bar{m}$ is the arithmetic mean particle mass, and σ_g is the geometric standard deviation.

Equation (1) will be assumed to represent the distributions of PuO_2 particles associated with the release circumstances considered in this report. The "fixed" values of σ_g and $\bar{d}_m$ are taken as 2.00 and 2μ respectively. These values are those given by Anderson (1964) as representative of the room temperatures release of PuO_2 associated with plutonium handling at Hanford. Since room temperature releases from metallic plutonium yield relatively fine particles compared to higher temperature releases (Stewart, 1963) this should be quite a conservative choice for $\bar{d}_m$. The range of $\bar{d}_m$ is 0.11μ to 4.2μ . This range includes the rather fine particles ($\bar{d}_m = 0.32\mu$) associated with the Rocky Flats lathe box fire mentioned in the previous section. The range of σ_g is 1.5 to 3.0. The fixed distribution and several distributions over the ranges of $\bar{d}_m$ and σ_g are shown in Figure 2.

The distributions represented by equation (1) must be modified when particle distributions in the enclosure leak paths are required. This modification is done by multiplying equation (1) by $K\delta^n$ where K is a constant and n is determined by the principle particle mode of transport. Since $\bar{m}$ involves an integration over $\bar{m}g(\delta)$, K will cancel when (1) is modified. The values of n are:

Settling; $n=2$
 Diffusion; $n=-3/2$
 Convection; $n=0$

The conservative assumption is made that the fixed value of n is $-3/2$ (diffusive transport) so that the smaller oxide particles are assumed to reach leak paths more readily and shift the log normal distributions present in the enclosure to a distribution in the leak paths characterized by a predominance of smaller particles which are less likely to be deposited in the leak paths.

3. Incidence Rate of Particles on Enclosure Leak Paths

A simple expression for the rate of particles entering leak paths may be obtained from considering the balance between particle formation and loss. The balance equation is:

$$\frac{dN_p}{dt} = G - L \quad (2)$$

where N_p is the number of air-borne plutonium oxide molecules present in the enclosure at any time, G is the rate of formation of the molecules and L is the rate of loss of the molecules. The formation rate is computed by assuming a total mass, M_t , of plutonium is divided into submasses which each have a mass M_1 and a surface area A_1 . During the release M_1 and A_1 are assumed to remain constant at their original value so that the formation rate is given by:

$$G = \frac{M_t}{M_1} A_1 K_g F_a \quad (3)$$

where K_g is the molecular release rate per unit area and F_a is the fraction of the released particles considered air-borne. K_g and F_a are assumed to remain constant in time. The particle loss rate is written:

$$L = R_h + R_w \quad (4)$$

where R_h is the rate of molecules entering leak paths and R_w is the rate of molecular deposition on interior enclosure surfaces. The forms of R_h and R_w are assumed to be: *

$$R_h = N_p A_h \text{ const}$$

$$R_w = N_p A_t f \text{ const} \quad (4-a)$$

* See Appendix I

where A_h is the total leak path entrance area, A_t is the total area of enclosure interior surfaces, and f is the fraction of the molecules which strike interior enclosure surfaces and are retained there. Implicit in equation (4) is the conservative assumption that if in one collision a particle does not stick to an interior enclosure surface, it must enter a leak path. Equations (3), (4) and (4-a) now are combined to give an expression for R_h on the assumption that a steady state has been achieved so that $dN_p/dt = 0$:

$$R_h = \frac{K_g M_t A_l F_a}{\left\{1 + \frac{A_t f}{A_h}\right\} M_l} \quad (5)$$

The fraction of the mass of air-borne particles from a release depends on the particle size distribution assumed. According to Mishima (1964) more than 99.9% of all PuO_2 particles, whose equivalent diameter is greater than 10μ , will in 10 minutes settle out of a turbulent room temperature aerosol in a 10 foot high chamber. It is thus assumed that whatever fraction of a particle distributions mass which is associated with particles of diameter less than 10μ is air-borne. Values of F_a for the particle distributions shown in Figure 2 are given in Table II:

TABLE II

PARAMETER RANGES FOR AEROSOL
PARTICLE DISTRIBUTIONS

$\bar{d}_m (\mu)$	σ_g	F_a
2.0	2.0	0.99
0.11	1.5	0.99999
0.11	3.0	0.999
4.2	1.5	0.981
4.2	3.0	0.78

The factor f may be evaluated from either of the following relations:

$$f = \frac{1}{1+K} \quad (6)$$

$$f = \frac{V_d}{V_t} \quad (6-a)$$

where K is the resuspension factor, V_d is the particle deposition velocity, and V_t is the transport velocity of particles normal to the deposition surface. -- The definition of K is:

$$K = \frac{\text{air-borne concentration (volume}^{-1}\text{)}}{\text{surface contamination (area}^{-1}\text{)}}$$

so that K has units of reciprocal length. Numerically K is the ratio of two numbers; the number of particles resuspended from a unit area of deposition surface into a unit volume which includes the unit surface area, and the number of particles remaining on the unit surface area after a resuspending disturbance. With this interpretation K may be used as indicated in equation (6).

According to Mishima (1964) the average value of K for PuO_2 aerosols is 4×10^{-8} and the proposed indoor resuspension factor with moderate activity is 5×10^{-5} . The latter number gives $f = 0.99995$.

The definition of V_d is:

$$V_d = \frac{\text{particle deposition rate per unit area}}{\text{particle concentration in gas immediately above surface}}$$

Mishima (1964) quotes experimental values of 3.2 cm/sec for 16μ and 29μ MMD size distributions and 0.1 to 0.3 cm/sec for submicron size particles. The values of V_t (cf., Schwendiman et al., 1963) for settling, diffusion and thermophoresis are

less than the corresponding values of V_d for PuO_2 particles $< 10\mu$ in equivalent diameter. Then from (6-a), f would be greater than one, implying 100% deposition on interior enclosure surfaces of all air-borne particles. The value of f is thus assumed to be unity in all calculations.

4. Description of Releases

a. Release Rates

The release rates vary over about 10 orders of magnitude depending on the crystalline phase and surface to mass ratio of the metal and the humidity, temperature, motion, and composition of the ambient gas. Stewart (1963) summarizes the existent data on release rates and from his compilation, a fixed release rate value of $10 \mu\text{ci}/\text{cm}^2\text{-hr}$ or $1.12 \times 10^{14} \frac{\text{Pu atoms}}{\text{cm}^2\text{-Sec}}$ is assumed. This value corresponds approximately to the maximum possible release rate under near static conditions which would give rise to the fixed particle distribution assumed ($\bar{d}_m = 2\mu, \sigma_g = 2.0$). The range of release rates for pure metal (Mishima, 1964) is a low of $1.5 \times 10^{-7} \mu\text{ci}/\text{cm}^2\text{-sec}$ for static room temperature oxidation in dry air to a high of $4 \times 10^4 \mu\text{ci}/\text{cm}^2\text{-sec}$ for dynamic release at δ phase temperatures (315°C to 452°C). The associated oxidation rate (from Mishima) is $11 \text{ mg PuO}_2/\text{cm}^2\text{-hr}$.

b. Area to Mass Ratio of Metal

The ratio A_1/M_1 in equation (5) is arbitrary and so what is felt to be a conservative fixed value and a conservative range of values is selected. The fixed value selected is $10 \text{ cm}^2/\text{g}$ which according to Mishima is typical of plutonium metal turnings. This implies fixed hourly oxidation and release rates of 11% and 0.17% respectively. The range of values is a low of 0.03 (which corresponds to a just critical pure metal sphere) to a high of 100 (which corresponds to plutonium "fines").

c. Total Mass Oxidized

The mass, M_t , in equation (5) is also arbitrary and again a conservative set of values must be assigned. The fixed value for M_t which is assumed is 202 kg. This number is selected since it has been estimated that the complete oxidation of this mass of plutonium in the test cell of the Rocky Flats Plant Critical Mass Facility would cause an overpressure of about 1.6 psi, which is the specified overpressure for leak testing of this test cell. In applying the model of restricted release to the Rocky Flats Plant test cell, which is done in the next section, it is thus very convenient to have the fixed room overpressure at just 1.6 psi. This number is felt to be quite conservative since it is a factor of almost 10 larger than the greatest amount of plutonium ever destroyed in any recorded accident. The limits on the mass are taken as 10 kg (which is about the critical mass of a bare plutonium sphere) and 1000 kg.

5. Leakage From the Enclosure

The leakage of air from the enclosure is assumed to occur via a number N_i of rough circular cross-section tubes of radius r_i . The tubes penetrate the walls perpendicularly and are straight except for a number, j , of kinks each consisting of two bends of equal and opposite radii of curvature and which have equal bend angles. The two bends which compose a kink are assumed to lie in the same plane and the spatial orientation of the kinks is assumed to be random. The radii of curvature, R_c , of the bends is taken to be of the order of the tube radii and to be given by an expression:

$$R_{ci} = K_c r_i$$

where K_c is a constant. The length of the leak paths is not assumed to be increased by the presence of kinks and so should be a minimum. The leak path model should thus be conservative.

The net airflow out of the room on the basis of the above model is:

$$Q = \sum_i V_i N_i \pi r_i^2 \quad (7)$$

where V_i is the velocity in a tube of radius r_i . V_i is assumed to be given by:

$$V_i = \frac{1}{\rho_{\text{air}}} \left[\frac{(\gamma-1) M_{\text{air}} g_c (P_o^2 - P_{\text{at}}^2) r_i}{2 \gamma R \bar{T} L \bar{f}(V_i)} \right]^{\frac{1}{2}} \quad (8^*)$$

where ρ_{air} is the density of air.

γ is the ratio of specific heats.

M_{air} is the mass per mole of air.

g_c is numerically equal to the acceleration of gravity.

P_o is the pressure inside the enclosure.

P_{at} is the atmospheric pressure outside the enclosure.

R is the molar gas constant.

$\bar{T} = (T_o + T_{\text{at}})/2$ is the average flow temperature.

T_o is the enclosure temperature.

T_{at} is the atmospheric temperature.

L is the tube length (all tubes are assumed to be of equal length).

$\bar{f}(V_i)$ is the average Fanning Friction Factor.

Equation (8) is derived by Knudsen and Katz (1958) in Chapter 7, of their book on fluid dynamics and is modified to include the effect of work done in an ideal gas adiabatic expansion.

* See erratum sheet

Equation (8) assumes:

- a. An average flow velocity over the entire tube length - This is a very realistic assumption for the short path length and low viscosities involved in the flow.
- b. The tube is horizontal - For a random distribution of tube directions in an enclosure wall this should be a reasonable assumption.
- c. The gas flow is adiabatic - This is a good approximation for laminar flow but for turbulent flow in small tubes (diameter of the order of a millimeter) it will probably overestimate the velocity by a factor of about two because of turbulent convective heat transfer.
- d. Average values of T and f are assumed along the length of the tube - This is a reasonable approximation since these quantities will not vary greatly over the tube length.
- e. The decrease of velocity associated with the leak path kinks is neglected - Pressure head losses associated with tube bends are usually small but for a path with several kinks at high flow velocities and high Reynolds number a significant error can occur. However since inertial deposition will be assumed to be negligible for turbulent flow this should be a very good approximation.
- f. The expression is valid for both subsonic and supersonic flow - The friction factors are not well known for supersonic flow and also stagnation pressure losses, which are not accounted for in equation (8, will start to become significant (Shames, 1962) so that equation (8 becomes somewhat inaccurate. It will however be shown in Part 5 of this section, that this inaccuracy will not significantly affect the leak path deposition probability.

g. The flow is one dimensional - This is a good approximation for the constant area circular cross-section leak paths assumed.

h. No "shaft work" is done.

The friction factor values outside the region $2000 < Re < 3000$ (where Re is the Reynolds number for fluid flow in a pipe) are taken from Knudsen and Katz (1958) and are:

$$f = 16/Re \text{ (Laminar flow in smooth or rough pipes; } Re < 2000) \quad (9-a)$$

$$f = 0.00140 + 0.125(Re)^{-0.32} \text{ (Turbulent flow in smooth pipes; } Re > 3000) \quad (9-b)$$

$$f^{-\frac{1}{2}} = 4 \log \left\{ \frac{2r_i}{e} \right\} + 2.28 \text{ (Fully turbulent flow in rough pipes; } \frac{r_i/e}{Re^{\frac{1}{2}}} > 0.005) \quad (9-c)$$

$$f^{-\frac{1}{2}} = 4 \log \left\{ \frac{2r_i}{e} \right\} + 2.28 - 4 \log \left\{ 1 + 4.67 \left[\frac{2r_i/e}{Re^{\frac{1}{2}}} \right] \right\} \quad (9-d)$$

(Transition flow in rough pipe; $Re > 3000$)

The quantity e is a roughness parameter which has the dimensions of r_i . Values of e for several materials are given by Knudsen and Katz.

The variation of f with Re in the range $2000 < Re < 3000$ is not known and so arbitrary models for f must be adopted here. For smooth tubes, a linear relation is assumed between the values $f(Re=2000)$ and $f(Re=3000)$ given by the two smooth pipe recipes above. The discontinuities in slope thus introduced are small enough that the required iterative

solution of equation (8 is easily accomplished for all Reynolds numbers. For rough tubes, the transition flow recipe is assumed initially to be valid in the region $2500 < Re < 3000$ and the laminar flow recipe to be valid in the region $2000 < Re < 2500$. For Reynolds numbers in the vicinity of 2500, the iterative solution of equation (8 will then oscillate between "laminar" flow ($Re < 2500$) and "turbulent" flow ($Re > 2500$). The solution of (8 is accomplished in these cases by using the linear average of the oscillations as a first approximation to V_i . If the value of Re associated with this first approximation is greater than some cutoff value, V_i is then found iteratively assuming equation (9-d is valid for all Re . If the first approximation to Re is less than the cutoff value, V_i is found by assuming equation (9-a is valid at all Re . The cutoff value must be less than 2500 since iterative solution of (8 in either branch causes Re to increase. A cutoff value of $Re = 1900$ was used in the calculations and led to acceptable physical results. It is estimated that values of V_i computed from equation (8 in the region $2000 < Re < 3000$ may be in error $\pm 25\%$.

The leakage paths are specified by the following parameters:

1. The size distribution of the leakage tubes.
2. The variation of the size distribution with enclosure overpressure.
3. The number of leak paths.
4. The number of bends in the tubes.
5. The bend angle distribution.
6. The bend radii of curvature distribution.

The choice of the r_i is made by utilizing whatever physical knowledge is available to estimate the characteristic leak path dimensions. It is usually impossible to specify an r_i distribution so an average model of r_i vs enclosure overpressure is assumed in any one leakage calculation. This average model

is then varied to account for any lack of conservativeness introduced by this assumption. Fixed variations of r_i with pressure and ranges of these variations are specified for the Rocky Flats Plant critical assembly cell in the next section.

The value of N_i is computed from equations (7 and (8 by using values of Q , P_{at} and T_o , obtained either from measurements of the leakage rate from the enclosure or from a specification of permissible leakage rates. This procedure gives values of N_i which ensures that the leakage model is "correct" at one overpressure. This N_i is assumed independent of pressure (cf. Chi and Kirstein, 1958) and is the N_i used in calculations of Q for a particular r_i vs enclosure overpressure model.

The limits set up for enclosure leakage rates refer to static overpressure in enclosures with small but numerous leak paths whose characteristic dimension is a few hundredths of a millimeter. Therefore in the evaluation of enclosure leakage rate limits it is not necessary to consider the effects of large characteristic dimension leak paths such as unsealed conduits or open doors. It is however interesting and important from a "maximum credible accident" standpoint to consider such leak paths, and so in the next section the maximum safe diameter of a single open straight pipe will be computed for the fixed release values. The maximum safe number of pipes of any diameter, less than the single pipe safe diameter, may also be readily obtained from the computations.

The specification of the leak path bends is also arbitrary and so fixed and extremal values are assigned to them. The specification for the Rocky Flats Assembly Room are given in the next section.

6. Deposition of Particles in Leak Paths

Of the oxide particles which enter leak paths only a certain fraction, F , will traverse them without being deposited

on the tube walls. The following deposition mechanisms are considered:

- a. Gravitational settling
- b. Brownian diffusion
- c. Thermal conduction
- d. Inertial impaction
- e. Turbulence

The settling mechanism is only important for oxide particles of greater than about one or two microns in diameter, while the diffusion and conduction mechanisms are significant for particles of less than about one tenth micron diameter.

The inertial mechanism increases in importance for laminar flow as the flow velocity increases and will usually be the dominant deposition mode for near sonic velocities. Turbulent deposition will usually dominate all other mechanisms for turbulent flow and for particles of less than one or two microns diameter. The turbulent deposition mode also increases in importance with increasing velocity. The deposition probabilities for the mechanisms assumes that if an aerosol particle is transported to a conduit wall it is sure to be retained there. This is the usual assumption made in studies of aerosol transport (cf., Postma, 1961; Browning and Ackley, 1962).

a. Gravitational Settling

The formula used for computing non-deposition due to settling (F_g) is according to Thomas (1958):

$$F_g = \frac{3}{2\pi}\beta\sqrt{1-(3/8\beta)^{2/3}} + \frac{2}{\pi}\sqrt{1-(3/8\beta)^{2/3}}\left\{\frac{3}{8}\beta\right\}^{1/3} + \frac{2}{\pi}\sin^{-1}\sqrt{1-\left\{\frac{3}{8}\beta\right\}^{2/3}} \quad (10)$$

F_g = non-deposition probability for settling

$\beta = Lu_t/r\bar{V}$

L = tube length (averaged over-all leak path directions)

r = tube radius

$\bar{V}$ = average velocity in tube

u_t = terminal settling velocity = $\frac{g\delta^2(\rho_p - \rho_g)K_m}{18\mu}$

g = gravitational acceleration

δ = particle diameter

ρ_p = particle density

ρ_g = gas density

μ = gas viscosity

K_m = Cunningham correction factor = $\left\{1 + \frac{2A\lambda}{\delta}\right\}$

λ = mean free path of gas molecules

$A = 1.26 + 0.4 \exp\left[-1.1\delta/2\lambda\right]$ (Davies, 1945)

Equation (10 assumes:

1. Parabolic velocity profile
2. Laminar flow
3. $Re_p = \rho_{air} V_i \delta / \mu \lesssim 0.1$
4. An incompressible infinite medium
5. Unaccelerated motion
6. Rigid spherical particles
7. No slip at the surface of the sphere.

Requirements (1 through (5 are either satisfied or contribute negligibly to the accuracy of equation (10). The asymmetry of the oxide particles will lead to an increased Stokes' Law resistive force but this will be compensated for by the diminished resistive force due to the failure of condition (7 to be satisfied (Fuchs, 1964). Equation (10 should thus adequately represent particle deposition by settling in circular tubes.

b. Brownian Diffusion

The problem of diffusive deposition of particles on the walls of circular tubes has been treated by several authors. The equations used here are due to Gormley and Kennedy (1949). For large deposition:

$$F_d = 0.819e^{-\alpha} + 0.097e^{-6.1\alpha} + 0.032e^{-16\alpha} \quad (11)$$

F_d = non-deposition probability due to diffusion

$$\alpha = \frac{3.66\pi DL}{Q}$$

Q = flow rate in tube = $\pi r_i^2 V_i$

D = diffusion coefficient = $\frac{RTK_m}{3\pi N_o \mu \delta}$

R = molar gas constant

T = absolute temperature

N_o = Avogadro's number

For small deposition, Gormley and Kennedy give the equation:

$$F_d = 1 - 1.09\alpha^{2/3} - 0.32\alpha + 0.0097\alpha^{4/3} \quad (11-a)$$

Schwendiman, et al., (1964) suggest the use of (11-a) for $F_d \geq 0.78$ and this convention has been adopted. Equations (11 and (11-a) assume:

1. Parabolic flow
2. Laminar flow
3. Flow velocity much greater than diffusive velocity
4. No tube entrance effects.

Conditions (1 through (3 are usually satisfied while condition (4 is usually not satisfied and will cause the equations to underestimate the deposition. This underestimate should however only be significant for particles of less than $10^{-3}\mu$ diameter (Fuchs, 1964).

c. Thermal Conduction (Thermophoresis)

According to Postma (1961) an expression for the change in concentration of particles in a circular tube as a function of the inlet gas temperature and the tube wall temperatures is:

$$F_c = \exp \left[- \frac{3K_g K_m (T_o - T_w)}{2(2K_g + K_p) T_w} \right] \quad (12)$$

where K_g = gas thermal conductivity

T_o = inlet absolute gas temperature

T_w = absolute wall temperature

K_p = particle thermal conductivity

F_c = non-deposition probability for thermal conduction

Equation (12) assumes:

1. The tube length to diameter ratio is ≥ 100
2. The quantity $\frac{5(f/2)^{\frac{1}{2}}(T_o - T_w)}{T_w} \ll 1$
3. The concentration profile is flat in the laminar sublayer
4. Turbulent flow exists.

These assumptions are ordinarily met for the conditions considered in this report when turbulent flow occurs.

d. Inertial Impaction

As particles traverse bends in the leak paths, a centrifugal force is developed which causes the particles to move radially outward. The radial motion will be opposed by a Stokes' law resistive force given by:

$$F = \frac{3\pi\mu\delta V_r}{K_m} \left[1 + \frac{3}{16} \frac{\delta \rho_g V_r}{\mu} \right] \quad (13)$$

where μ = the viscosity of air

δ = the particle diameter

V_r = the particles radial velocity

ρ_g = the density of air

K_m = Cunningham slip correction factor

The term is the parenthesis in a correction factor given by Oseen (1927) which partially accounts for the neglect of inertial forces implicit in the derivation of Stokes' Law. The centrifugal force is assumed given by:

$$F = \frac{mV_t^2}{R_c} \quad (13-a)$$

where V_t is the tangential velocity of the particle traversing the bend, R_c is the average radius of curvature of particles in the bend and m is the particle mass. The motion in the bend cannot be considered uniform and so the equation for radial motion using (13 and (13-a is:

$$m \frac{dV_r}{dt} = \frac{mV_t^2}{R_c} - \frac{3\pi\mu\delta V_r}{K_m} \left[1 + \frac{3}{16} \frac{\delta \rho_g V_r}{\mu} \right] \quad (14)$$

It is assumed that the particles are sufficiently entrained in the flow that the particles tangential velocity in traversing the bend is constant or $V_t = V_i$ [equation (8)]. Also the flow velocity profile is assumed to be unaltered as the fluid goes around the bend. Thus equation (14 may be solved for the radial displacement, Δr , of a particle subject to the boundary condition $V_r = 0$ at $t=0$ or:

$$\Delta r = \frac{1}{K_2} \ln \left| \frac{p \exp \left[-K_2 (p-q) R_c \theta / V_i \right] - q}{p-q} \right| + \frac{p R_c \theta}{V_i} \quad (15)$$

where p and q and the roots of the equation:

$$K_2 V_r^2 + K_1 V_r - K_0 = 0 \quad (15-a)$$

$$\text{and } K_2 = \frac{18^0 g^K p}{K_m \rho_p \delta}$$

$$K_1 = \frac{18\mu}{K_m \rho_p \delta^2}$$

$$K_0 = V_i^2 / R_c$$

$$K_p = 0.1875$$

$$\rho_p = \text{particle density}$$

The time has been related to the bend angle, θ , by

$$t = \frac{R_c \theta}{V_i} \quad (15-b)$$

The inertial non-deposition probability, F_b , is computed from equation (10 with $\beta = \frac{\Delta r}{r}$, where r is the tube radius.

If more than one bend per leak path is assumed, F_b in equation (19-a:

$$F_b = \{F_{bo}\}^j \quad (16)$$

where j is the number of bends and F_{bo} is the single bend non-deposition probability. This is a reasonable assumption since it may be assumed that the several kinks in a tube are randomly oriented with respect to each other.

A resume' of the assumptions made in deriving equation (15) is:

1. An average bend curvature radius for the particles - The effect of this approximation is estimated in the next section.
2. Parabolic velocity flow profile which is unaltered during flow around the bend.
3. No tangential particle slippage in traversing the bend.

e. Turbulence

Above a flow Reynolds number of 2500 turbulent flow is assumed to occur and turbulent deposition and thermal conduction are conservatively assumed to be the only deposition modes. Using the idea of the deposition velocity (V_d) previously discussed, a general expression for the non-deposition probability in circular constant cross-section tubes is given by:

$$F = \exp\left[-\frac{4V_d L}{V_i d}\right] \quad (17)$$

V_i = average flow velocity

L = tube length

d = tube diameter

F = non-deposition probability

The ratio V_d/V_i is called the mass transfer coefficient. For turbulent deposition, Schwendiman, et al., give the following expression for V_d/V_i (for air at 25°C):

$$V_d/V_i = 14.14 \left(\frac{d^{0.84} f \delta^2 \rho_p V_i^2 \rho_g}{13.5 \rho_p \delta^2} \right)^{2.015} \quad (18)$$

$$\mu^2 \left[1 + \frac{\mu}{\rho_p} \right]$$

where the meaning of terms is the same as in equations (8, (13 and (17.

When equations (17 and (18 are combined to give a turbulent non-deposition probability, F_t , the limit of F_t as $d \rightarrow \infty$ is zero. To avoid this physically impossible situation the criteria of Friedlander and Johnstone (1957) is used. This criteria is that for particles whose reduced stopping distance, S^+ , is greater than 30, the expression for V_d/V_i is given as:

$$\frac{V_d}{V_i} = \frac{f}{2} \quad (18-a)$$

The reduced stopping distance, which physically is the distance a particle will travel before stopping when acted on by only a Stokes' Law type resistance force, is given by:

$$S^+ = \frac{f \delta^2 \rho_p V_i^2 \rho_g}{40 \mu^2} \quad (18-b)$$

This criteria ensures that under most circumstances of interest in this report, supersonic flow velocities will not effect the deposition probabilities appreciably. For example, for just sonic, turbulent flow of air in rough tubes whose asymptotic value of f is 0.018, equation (18-a will apply for all particles greater than 0.155μ in diameter. Since equation (9-c shows that at high Reynolds numbers, f is independent of Reynolds number, the applicability of (18-a implies deposition probabilities will not be much affected by changes in supersonic flow velocities. Equations (17, (18, (18-a and (18-b are assumed to specify the turbulent deposition probability..

They assume:

1. Turbulent flow ($Re > 2500$)
2. Constant cross-section circular tubes
3. All particles which approach within one stopping distance of the tube wall are removed from the flow.
4. The initial velocity used in calculating the stopping distance is the r.m.s. velocity of turbulent fluctuations in the direction normal to the wall.

7. The Total Deposition Probability

The net probability for the non-occurrence of an event when there are several independent and simultaneous ways for the event to occur is the product of the several independent non-occurrence probabilities. It is assumed here that all the non-deposition mechanisms are independent so that the expression used to compute the total deposition probability, F , is:

$$F = F_g F_d F_c F_t F_b \quad (19)$$

For laminar flow, equation (19) is assumed to be:

$$F(Re < 2500) = F_g F_d F_b \quad (19-a)$$

and for turbulent flow:

$$F(Re > 2500) = F_c F_t \quad (19-b)$$

The assumption of independent deposition modes is usually quite good. When equation (19-a) applies, F_g and F_d are independent since they are important for particles in different size ranges. F_g and F_b are independent because of the random orientation of the kinks and the difference in velocity ranges in which they are significant. F_d and F_b are independent because they apply

to different size particles and are important in different regions of flow velocity. When equation (19-b) applies F_c and F_t are independent since F_c is only important for very small particles ($\delta < 10^{-2} \mu$).

8. The Free Release Source Strength Equation

The final expression used in computing the free release source strength is:

$$S = \sum_{i,j} R_{hi,j} \bar{F}_{i,j} \quad (20)$$

$$\text{where } \bar{F}_{i,j} = \int_{\delta_{\min}}^{\delta_{\max}} F_{i,j}(\delta) g(\delta) \delta^y d\delta \quad (20-a)$$

The particle distribution, $g(\delta)d\delta$, is given by equation (1). The exponent y is equal to $n+2$, with the value of n for the different modes of particle transport to the enclosure walls given in Part 2 of this section. $\bar{F}_{i,j}$ represents the average non-deposition probability for tubes of radius r_i and containing j bends per leak path. $R_{hi,j}$ is the rate of incidence of particles on leak paths of the i,j th type as given by equation (5 with $A_h \equiv A_{hi,j}$ being the total entrance area of i,j type leak paths. The integration limits in (20-a) are $10^{-3} \mu$ and 10μ according to the discussion of particle size distribution in section C-2. A computer program to compute S for different leak path and release models has been written in FORTRAN II for the A.S.I. 210.

9. The Relation Between Source Strength and Air-Borne Particle Concentration

A conservative estimate of the conversion of source strength S to maximum air-borne concentration χ is obtained from the work of Hilsmeier and Gifford (1962). The following assumptions are made:

a. Point Release Occurs With Volume Source Dilution

This means that the cross-sectional area of a point source plume normal to the wind direction is increased by a factor which is of the order of the source area normal to the wind direction. The normal source, A_n , will be assumed to be the minimum vertical cross-section area which includes the vertical symmetry axis of the enclosure. The multiplicative factor will be conservatively taken as 0.5 (Gifford, 1961). The point source concentration, χ_o , is thus divided by a factor $(1+0.5A_n/\sigma_y\sigma_z)$ where σ_y^2 and σ_z^2 are the horizontal and vertical dispersion coefficients respectively.

b. Ground Level Release Occurs

This assumption may be shown to overestimate the actual maximum concentration by a factor of about 500 for releases from a height above the ground equal to half the test cell height.

c. No Particle Sinks are Present

This means that no surface deposition or washout of particles is assumed. This assumption is fairly realistic and should not overestimate the actual maximum concentration by more than 10 to 20%.

d. Moderately Stable "Type F" Atmospheric Conditions Exist

The probability here is roughly 0.15 for such conditions to exist at the Rocky Flats Plant site. The wind velocity, $\bar{u}$, associated with such conditions is about 2 m/sec. This assumption minimizes the spread of the plume and can overestimate χ by as much as a factor of 40 (see Figure 3).

e. The Concentration is Measured on the Plume Axis

The probability of a person being just downwind from the facility when a release occurs is slight but this assumption is necessary to obtain an estimate of the maximum possible concentration associated with a release.

f. A Source Strength Which is Constant in Time

Based on the above assumptions, the maximum possible value of the quantity $\bar{u}_{\chi_0}/S$, according to Hilsmeier and Gifford, is 4 (cf. Figure 3). This value occurs at a distance of 10 meters from the source. The relation between χ and S is then:

$$\chi = \frac{2S}{\left[1 + A_n / \pi \sigma_y(10) \sigma_z(10) \right]} \quad (21)$$

where χ and S must have compatible MKS units. From extrapolation of the curves prepared by Hilsmeier and Gifford, $\sigma_y(10)\sigma_z(10)=0.15\text{m}^2$ so that (21) becomes:

$$\chi = \frac{2S}{1 + 2.12A_n} \quad (21-a)$$

D. APPLICATION OF RESTRICTED RELEASE THEORY

1. The Rocky Flats Plant Critical Assembly Test Cell

The theory outlined in the previous section is now used to evaluate the leakage rate limits of the Rocky Flats Plant Critical Mass Facility test cell. The leakage rate limits specified for this test cell are less than 3.0% total volume leakage in 6 hours and no greater than 0.5% total volume leakage in any 1 hour at an overpressure of 1.6 psi. The Rocky Flats Plant cell has been leak tested and found to conform to these limits. Physically, the test cell is a rectangular parallelepiped with concrete walls, roof, and floor. The cell interior is 35 feet long, 32 feet wide, and 32 feet high. The north wall, which is between the experiment and the experimenters, is 5 feet thick while the other walls are 4 feet thick. The cell floor is 8 in. thick and the roof is about 2 feet 2 in. thick. The building was constructed of

cast-in-place blocks with the walls being done in three concrete pours and the roof and floor in single pours. The walls, floor, and roof all contain a 12 in. rebar mesh recessed 3 in. from each surface in the walls and ceiling and about 1 in. from the floor surfaces. The nominal rebar size is number 8 (8/8 in. diameter). All the interior and accessible exterior surfaces have received several coats of paint or other sealants to reduce leakage through the concrete itself.

The south cell wall contains a pair of 8'6" x 4'4" x 5" blast doors and an associated cement filled 3'6" thick shield door while the north wall contains a similar single blast door. The walls and floor of the cell, especially the north wall, contain utility penetrations for water, electricity, air, or experimental solutions. The cell is provided with its own heating, ventilation, and air conditioning system which necessitates a 12" diameter intake air pipe penetrating the west wall and a 10" diameter exhaust air pipe penetrating the floor. All of the above mentioned cell penetrations are minor leak sources if the doors are properly closed and no valves are left open thus implying most of the cell leakage occurs through the walls themselves.

2. Variation of Test Cell Temperature With Amount of Plutonium Oxidized *

In order to avoid the great difficulty involved in any rigorous calculation of space and time variation of temperature within the cell a simple calorimetric calculation is done making the following assumptions:

- a. The cell temperature has no spatial variation and a steady state always exists in the cell between heat sources and sinks.
- b. The cell heat source is the plutonium plus oxygen reaction whose reaction heat is 1058 cal/g-Pu (Hilliard, 1963).
- c. Four potential heat sinks exist in the test cell:

- (1). Ten tons of iron of specific heat $0.12 \text{ cal/g-}^{\circ}\text{K}$.
This corresponds roughly to the mass of the experimental apparatus which exists in the test cell.
- (2). A shell of concrete 1 centimeter thick and having the area of the interior surfaces of the cell. This gives a heat sink of about twenty tons with a specific heat of $0.20 \text{ cal/g-}^{\circ}\text{K}$.
- (3). One and one-half tons of air with a specific heat at constant volume of $0.17 \text{ cal/g-}^{\circ}\text{K}$.
- (4). Heat loss through leak paths by convection.

On these assumptions the relation between the cell temperature, $T_o (^{\circ}\text{K})$, and the mass of plutonium oxidized, M_t (moles), is:

$$T_o = TCON M_t + 293 \quad (22)$$

where TCON is a constant specifying the type of thermal equilibrium assumed. Values of TCON for several types of equilibrium are shown in Table III. The assumed fixed and extremal values of this parameter also appear in Table III.

TABLE III

THERMAL EQUILIBRIUM TYPES	
Heat Sinks	TCON ($^{\circ}\text{K/mole}$)
Air, iron, concrete and leak paths	0.0239 (minimum)
Air, iron, concrete	0.0511 (fixed)
Air, concrete	0.0652
Air, iron	0.191 (maximum)
Air	1.090

The conversion of cell temperature to cell pressure is done using the perfect gas law. At the elevation of the Rocky Flats Plant this gives:

$$P_o (\text{dynes/cm}^2) = 2.75 \times 10^3 T_o$$

3. Model of Test Cell Leak Paths

a. Pressure Dependence of Leak Path Diameters

The average model of leak path radii vs external pressure is taken from the work of Chi and Kirstein (1958) who have studied the factors which determine the width and spacing of flexural cracks in reinforced concrete. The differences in rebar pattern, size, heat generated in cement setting, and surface treatment of the test specimens of Chi and Kirstein and the blocks of which the test cell is constructed as well as the assumption that the leak paths have the surface dimension through their entire penetration of the concrete make this only an order of magnitude model. It is suitable however as a starting point for studying the effect of leak path size variation on the source strength. The dependence of r_i on external pressure is derived in two steps: First, the following expression is written (where $r_i \equiv \bar{r}$ since all leak paths are assumed to have the same radius):

$$\bar{r} = a\sigma + b \quad (24)$$

where σ is the stress in a rebar. The fixed values of a and b are obtained from Chi and Kirstein and refer to surface cracks in a 132" x 20.94" x 6" test specimen containing two number nine rebars. The ranges of a and b values are also estimated from Chi and Kirstein and are shown along with the fixed values in Table IV.

TABLE IV

LEAK PATH SIZE THERMAL RESPONSE PARAMETERS		
	$a(\text{cm}^3/\text{dyne})$	$b(\text{cm})$
Fixed	5.57×10^{-12}	-3.55×10^{-3}
Maximum	1.37×10^{-11}	-1.42×10^{-3}
Minimum	2.225×10^{-12}	-5.68×10^{-4}

The second step is the derivation of an expression for σ vs overpressure. This expression is obtained on the assumption that each rebar acts independently of each other

and of the concrete in which it is imbedded. The expression is derived from the equation of bending of a beam (on the assumption that both ends are fixed) and is:

$$\sigma_{\max} = \frac{1}{12} \frac{Wlc}{I} \quad (24-a)$$

where $\sigma_{\max}$ is the maximum stress in the rebar

W is the uniformly distributed total load

l is the rebar length

c is the half depth of the rebar

I is the moment of inertia of the area of a rebar section about the neutral axis. The neutral axis is assumed to be a section diameter.

For a cylindrical rebar of diameter d, $I = d^4 \pi / 64$, $W = ld \Delta p$ and $c = d/2$ where Δp is the test cell overpressure. Thus 24-a may be written:

$$\sigma_{\max} = \frac{8}{3\pi} \frac{l^2}{d^2} \Delta p \quad (24-b)$$

For The Rocky Flats Plant test cell with $d=1"$ and $l=30'$, (18-b gives:

$$\sigma_{\max} = 1.1 \times 10^5 \Delta p \quad (24-c)$$

Equation (24-c is combined with equation (24 and the Table IV values of a and b for the calculation of $\bar{r}$ vs overpressure for the test cell.

b. The Model of Inertial Parameters

The fixed values and range of inertial parameter values are given in Table V:

TABLE V

LEAK PATH INERTIAL DEPOSITION PARAMETERS			
	Bends per leak path	Bend angle (radians)	Bend radius of curvature
Maximum	10	π	$\bar{r}$
Fixed	1	$\pi/2$	$3\bar{r}$
Minimum	0	-	-

The fixed value of one ninety degree bend should be a conservative estimate for obtaining the inertial deposition properties of any possible leak path in the test cell. The maximum values of Table V are felt to more nearly approach the physical situation being simulated here.

c. The Leak Path Roughness

The primary composition of leak path walls is assumed to be concrete whose range of e values according to Knudsen and Katz is 0.0305 cm to 0.305 cm. Secondary leak paths may be through metal pipes ($e \approx 0.0045$ cm) or around rubber gaskets ($e \approx 0.01$ cm). The fixed e value and range of e values are shown in Table VI:

TABLE VI

LEAK PATH ROUGHNESS VALUES	
Minimum	$e = 0$ cm
Fixed	$e = 0.0305$ cm
Maximum	$e = 0.305$ cm

4. Results and Their Interpretation

a. Effect of Varying Release Description Parameters; Fixed Leakage Rate

Figures 4 and 5 show the variation of the free release source strength with, respectively, particle size distribution and mode of particle transport to enclosure walls. In Figures 4-a and 4-b the mass median diameter of the assumed particle distribution is plotted vs source strength for three values of σ_g and two plutonium masses. For the fixed situation of 202 kg of oxidized plutonium (Figure 4-a)

and $\bar{d}_m = 2\mu$; $\sigma_g = 2$, it is seen that varying σ_g can increase S by about two orders of magnitude (10^9 to 10^{11}) while varying $\bar{d}_m$ can increase S by almost three orders of magnitude (10^9 to 10^{12}). Other observations from Figure 4 are:

- (1). For small particles, the effect of particle dispersion variation is negligible. This is due to the relative insensitivity of the non-deposition probability of small particles to changes in particle size.
- (2). For sufficiently large and sufficiently mono-disperse particle distributions, no particles can escape the test cell. Thus the $\bar{d}_m = 4\mu$; $\sigma_g = 1.5$ distribution leads to perfect containment of the aerosolized plutonium.
- (3). Source strengths associated with laminar flow leakage from the enclosure, which occurs when 50 kg of plutonium is oxidized (Figure 4-b), are less affected by particle distribution variation than are the turbulent flow leakage source strengths which accompany the 202 kg oxidation.

The dependence of source strength on the mode of particle transport to interior enclosure surfaces is shown in Figures 5-a and 5-b where the source strength is plotted vs the exponent y of equation (20-a). As previously mentioned $y=1/2$ refers to diffusive transport, $y=2$ to convection, and $y=4$ to pure settling. All intermediate values of y refer to mixtures of these transport mechanisms. The curves of Figure 5 are parametric in the particle size distributions. Figure 5-a refers to oxidation of 202 kg of plutonium and Figure 5-b to oxidation of 50 kg of plutonium. For the fixed particle distribution with $\bar{d}_m = 2\mu$ and $\sigma_g = 2$, Figure 5-a shows that the fixed assumption of $y=1/2$ may overestimate the source strength by over four orders of magnitude (2×10^9 to 10^5). Other features of interest which are shown by Figures 5 are:

- (1). For all particle distributions and masses oxidized, the fixed assumption of $y=1/2$ is conservative.
- (2). The turbulent flow source strength, associated with the 202 kg oxidation, is more affected by variation of y than are the laminar flow source strengths accompanying the oxidation of 50 kg of plutonium.
- (3). Source strengths for large particle distributions are affected more by variation of y than are those for small particle distributions.

b. Effect of Varying Release Description Parameters; Variable Leakage Rate

Figures 6 and 7 respectively show the source strength variation with the thermal equilibrium type assumed in the enclosure and with the mass of oxidized metal. The variation of source strength with TCON can be significant since an increase in TCON increases the flow velocity, both through an increase in leak path radius and an increase in the pressure, and also increases R_H [equation (5)] through the increase in leak path radius. In Figure 6, values of TCON (see Table III) are plotted vs source strength for several different masses of plutonium oxidized. It is seen that for the fixed mass of 202 kg, the source strength may increase to 7.6×10^9 plutonium atoms/sec for the maximum TCON considered. The maximum TCON is not assumed to be the value for thermal equilibrium with air alone since this would require a release rate of about 10^5 $\mu\text{ci}/\text{cm}^2\text{-hr}$ which would yield a very coarse particle distribution (Stewart, 1963). Such an extreme type of release would represent the simulation by the model of explosive dispersion. *

The effect on the source strength of the mass oxidized for the fixed release conditions and leak path models is shown in Figure 7. The minimum mass oxidized of 10 kg is

* See Appendix II

shown to be perfectly contained by the enclosure while the fixed mass leads to a 2×10^9 source strength. The maximum mass of 1000 kg is shown to give rise to a source strength of about 8×10^{11} atoms/sec or about two and one half orders of magnitude greater than the fixed value. The structure of the curve in Figure 7 is explainable as follows: For small masses (<15 kg) the flow velocity is slow enough that diffusion and settling remove all the aerosol particles in the leak paths. As the mass increases, the flow velocity increases so that settling and diffusion become less effective but inertial impaction increases in importance. Thus between 20 kg and 55 kg, the source strength at first increases and then decreases. Between 55 and 70 kg, the transition from laminar to turbulent flow occurs. Above 70 kg, turbulent deposition and thermal conduction are assumed to be the only deposition mechanisms in the leak paths so that an initial and physically unreal rise occurs. Above 100 kg, turbulent deposition starts to become quite effective and a decrease in source strength occurs up to about 450 kg. Above this, the increase in enclosure overpressure and leak path radius causes equation (18-a for V_d/V_i to become applicable for the bulk of the particles involved in the release. The turbulent non-deposition probability then begins to tend to unity with an associated rise in source strength. The trends outlined above appear in all of the graphs involving a variation of S with a parameter which increases the flow velocity (cf. Figures 6, 8 and 11).

The release is also specified by the values assumed for the plutonium release rate and area to mass ratio. Since these quantities appear linearly in equation (5 and do not appear in equation (20-a, S is directly proportional to them both. Thus the value of S obtained from maximizing k_g for otherwise fixed conditions is $2 \times 10^9 \times \frac{4 \times 10^4}{10}$ = 8×10^{12} atoms/sec, while the value for A_1/M_1 is $2 \times 10^9 \times \frac{100}{10}$ = 2×10^{10} atoms/sec.

c. Effect of Varying the Leak Path Model

The effect of varying the leak path size for otherwise fixed leak path specifications and fixed release conditions is shown in Figures 8-a and 8-b. The fixed value and ranges of values for $\bar{r}$, as specified by a and b in equation (24 (cf. Table IV), are shown in Figures 8. It is seen in Figure 8-a that varying the leak path size may increase S by about one and one half orders of magnitude (2×10^9 to 7×10^{10}). The number of leak paths associated with $\bar{r}$ are shown in Figure 8-b. For the fixed radius of 6.6×10^{-2} cm the number is 23, while for the minimum radius of 2.7×10^{-2} cm the number is 650. The latter number is more physically representative of the number of leak paths which actually exist while the fixed radius selected represents the physical dimensions expected of the leak paths. The smaller numbers of leak paths actually computed for the fixed radius represents the attempt of the model to account for the constriction of the leak path entrances due to the several surface treatments received by the interior and exterior surfaces of the test cell. The model thus neglects any particle deposition or flow velocity reduction associated with the entrance constriction but underestimates the number of possible leak paths for the particles. Figure 8-a may be used to obtain the source strength for other masses oxidized at a given radius as follows:

- (1). Find S at a new radius given by $\bar{r} = \bar{r}(202 \text{ kg})M_t/202$ from Figure 8-a.
- (2). Multiply this new S by $M_t/202$.

The effect on S of changes in the inertial parameters appears in Figures 9. The increase of S possible as the inertial parameters change from fixed to extreme values (see Table V) is two orders of magnitude (1.7×10^8 to 1.8×10^{10}). Figures 9 also show:

- (1). The most pronounced variation of S is with the number of bends per leak path (Figure 9-a). Here S changes by almost five orders of magnitude over the range ($n=0$ to 10) of values considered.
- (2). S decreases as R_c increases (cf. Figure 9-b). This is because of the increased time needed to traverse the bend for larger radii of curvature. This effect, for R_c in the range considered, is larger than the opposing effect of a decreasing centrifugal force as R_c increases.
- (3). The maximum possible error due to assuming a constant radii of curvature for the particles may be obtained from Figure 9-b by finding S at $R_c \pm \bar{r}$. The error increases as R_c decreases and the greatest value in the range of R_c considered is a factor of 13.6 at $R_c=1$. At the nominal R_c value of 3, the factor is 2.5.
- (4). The variation of S with θ (Figure 9-c) is at first slight when "straight through" penetration of the tube is possible. As θ further increases S decreases more rapidly until the effect starts to saturate at about $\pi/2$. Above $\pi/2$ sufficient particles are sure to be deposited that a further increase in the particle "sink" (i.e. a sharper bend) is not as effective.

The leak path roughness affects the source strength only if turbulent flow occurs. The source strength is affected in two opposing ways by changes of e . First, increases in e decrease the flow velocity necessitating more leak paths and hence increasing S . Second, increases in e increase f so that for particles whose mass transfer coefficient is given by equation (18-a), the non-deposition probability will be decreased. It should be noted that the product fV_i^2 remains constant when V_i is changed by changing e . Thus for particles whose mass transfer coefficient is

given by equation (18, changes in e will not change the turbulent non-deposition probability. Also the criteria equation (18-b for determining the appropriate mass transfer coefficient recipe is unaffected by changes in e so that as e is varied for a given particle distribution the same fraction particles is affected by equations (18-a and 18-b respectively. The source strength change as e is changed, plotted for three masses, is shown in Figure 10. For the fixed 202 kg mass, the increase in S at extreme e values over that at the fixed e values (see Table VI) is only about a factor of two. As e increases, the flow may be forced to become laminar (as indicated in Figure 10). Above this critical value of e no S values are computed since changes in e only affect turbulent flow.

d. Effect of Varying Single Leak Path Size

In Figure 11-a the source strength variation for two masses is shown as a function of the size of a single straight leak path. Fixed release and leak path conditions are assumed except that the leak path radius is assumed pressure independent. The range of leak path radii considered is 10^{-3} cm to 10^3 cm. Below 5×10^{-3} cm no particle leakage occurs and from 5×10^{-3} cm to 2×10^{-2} cm, 202 kg source strengths are greater than 50 kg source strengths. From 2×10^{-2} cm to 2×10^{-1} cm, 50 kg strengths exceed 202 kg strengths while above 2×10^{-1} cm the 202 kg strengths always exceed the 50 kg strengths. The cross-overs of the source strengths result from the different velocities associated with the same size leak path when different plutonium masses are oxidized. Figure 11-a may be used to compute the net source strengths (for oxidation of 50 kg or 202 kg plutonium) associated with leakage through any combination of straight leak paths using the relation $S = \sum_i N_i S_i$. Figure 11-b shows the volume leakage percent per second of air from the test cell. It is seen that if this leakage rate could be sustained for

202 kg oxidation the entire air content of the cell would escape in one second through a single 30 cm radius leak path. The S associated with a 30 cm radius leak path is 7×10^{15} atoms/sec.

E. CONCLUSIONS

According to the method of analysis outlined in the introduction, namely assuming a set of accidents in which the release variables are individually maximized and from this set selecting a maximum credible accident, the maximum credible source strength for 1/2 v/o per hour leakage from the Rocky Flats Plant Assembly Room is 8×10^{12} Pu atoms/sec. The maximum safe source strength * is 1.23×10^{14} Pu atoms/sec which implies, since the release model assumes source strength is proportional to enclosure leakage rate, that the assembly room leakage rate could safely be increased a factor of 15.4. The model also shows that under fixed conditions a single straight leakage path as large as 3.5 cm in radius ** can exist in the Assembly Room walls before the plutonium air-borne concentration limits are exceeded (cf. Figure 11-a). Such a leak path would, according to the model, cause the entire Assembly Room to be evacuated in a little over two minutes if the initial release conditions could be sustained (see Figure 11-b).

The release model also indicates which variables are most significant in evaluating the potential hazard from an uncontrolled release of plutonium inside an enclosure. Table VII summarizes the principle fixed release and leak path assumptions and gives the relative importance of the assumptions to the potential release hazard as well as an estimate of their conservativeness. It is seen from Table VII

* See Appendix III

** See Appendix I

TABLE VII
EVALUATION OF FIXED RELEASE ASSUMPTIONS

<u>Fixed Assumptions</u>	<u>Relative Conservativeness of Assumptions</u>	<u>Factor By Which Extremizing This Variable Can Increase Source Strength</u>
Leak path radii and their pressure variation is that of cracks in reinforced concrete	Realistic assumption for the reinforced concrete test cell	4.2×10^1
Leak path roughness that of concrete	Realistic assumption	2
One 90° bend per leak path for laminar flow	Very conservative for leakage through 4 feet of painted concrete	10^2
Only turbulence and thermal conduction cause particle deposition in turbulent flow	Very conservative - neglects inertial deposition and settling in turbulent flow	-
Particle distribution with $\bar{d}_m = 2\mu$ and $\sigma_g = 2$	Conservative unless unoxidized plutonium has area to mass ratios of greater than 10	5×10^2
Area to mass ratio of plutonium metal equal to 10	Quite conservative - refers to lathe turning size	10^1
Release rate of plutonium is $10 \mu\text{ci}/\text{cm}^2\text{-hr.}$	Conservative unless a high flow rate of air is incident on the burning material	4×10^3

TABLE VII
EVALUATION OF FIXED RELEASE ASSUMPTIONS
(concluded)

<u>Fixed Assumptions</u>	<u>Relative Conservativeness of Assumptions</u>	<u>Factor By Which Extremizing This Variable Can Increase Source Strength</u>
202 kg of plutonium involved in the release	Very conservative - about 10 times the amount of plutonium ever destroyed in any recorded accident	4×10^2
Diffusion always transports the released particles to the enclosure walls.	Always conservative; may overestimate S by as much as four orders of magnitude	1
A thermal equilibrium is * achieved in the enclosure during the release. The source is the $\text{Pu} + \text{O}_2 \rightarrow \text{PuO}_2$ heat of reaction and the three sinks are:	A realistic assumption unless the entire release occurs in a few seconds or less	3.8×10^2
1. The air in the enclosure		
2. The test equipment in the enclosure		
3. The first 1 cm of the interior concrete surfaces of the enclosure.		

* See Appendix II

that the most important variables are the release rate, the particle distribution, and the type of thermal equilibrium. Maximizing the release rate will maximize the thermal over-pressure but will generally result in a very coarse particle distribution which is easily filtered out by the leak paths. The principle hazard is a release rate of sufficient magnitude to breach the enclosure walls and explosively disperse plutonium via the resulting shock wave.

An operational criteria for enclosures handling plutonium is thus to continuously exhaust the enclosure via a series of baffled, fireproof, absolute filters. The baffle-filter combination serves to attenuate and filter the shock wave from any explosion involving plutonium while the continuous exhaust mode of operation prevents any static build up of pressure associated with long term plutonium oxidation.

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APPENDIX I

THE ENTRAINMENT OF AEROSOL PARTICLES IN AN AIR FLOW

The assumption made in equation (4-a) is that the fraction of aerosol particles, generated within an enclosure in which an overpressure exists, which reach leak path openings is the ratio of the leak path entrance area to the total enclosure interior area. An alternate procedure is to compute the fraction of the enclosure air which leaks in a given time and to estimate the fraction of the released aerosol particles which are entrained in this emitted air. The only requirement is that the procedure adopted is conservative under the range of release conditions assumed. The model used assumes an entrainment fraction of about 5.2×10^{-5} which is independent of the flow rate of air out of the enclosure. Some applicable data to evaluate this entrainment efficiency is that of Cheever (1961), whose experiments were discussed briefly in section B-2. Cheever finds $5.5 \times 10^{-4}\%$ of the plutonium volatilized in a glove box which was exhausted at a rate of 5 cfm was collected on the glove box filters. The average plutonium oxide particle size was 0.016μ which corresponds to a mass median diameter of 0.74μ if the count median and average diameter are taken as equal. This implies an average entrainment efficiency for these particles of 5.5×10^{-6} or, taking entrainment efficiency as proportional to particle mass, an entrainment efficiency of 1.68×10^{-6} for 0.11μ MMD particles. Then assuming the entrainment fraction is proportional to flow rate a flow of 154.7 cfm is required before the entrainment factor of 5.2×10^{-5} is exceeded for the smallest particles considered in the report. For the fixed size distribution of 2μ MMD, the maximum flow rate would be 9.77×10^6 cfm. According to the model this flow rate is associated with the leakage from a single straight 20 cm radius pipe and so the model should be conservative for fixed releases and pipes less than 20 cm in radius.

APPENDIX II

THERMAL EQUILIBRIUM IN THE ROCKY FLATS PLANT ASSEMBLY ROOM

In order to test the conservativeness of the assumptions made in evaluating the type of thermal equilibrium actually attainable in the Rocky Flats Plant Assembly Room the following experiment was performed: The Assembly Room was sealed and the room heating system turned on. The heating system controls were adjusted so that a continuous heat input was sustained. (The heating system is rated at 271,000 Btu/hr which corresponds to the heating rate associated with a fixed release whose release rate is 2.92×10^3 $\mu\text{ci}/\text{cm}^2\text{-sec}$ or a factor of only 13.7 less than the maximum release rate considered.) The room overpressure created by the heat input was measured with an oil manometer (oil density $0.82 \text{ g}/\text{cm}^3$) and the room temperature was read from a thermocouple thermometer observed over the Assembly Room closed TV circuit. The atmospheric temperature was obtained from the Rocky Flats Plant Health Physics thermograph records. The experiment was terminated after three hours with the following results:

- A. The temperature difference between the room and the atmosphere increased 11°F in the first 30 minutes and 19°F overall.
- B. No measurable overpressure was observed.

This experiment demonstrates the efficiency of the Assembly Room heat sinks and also the rapidity with which a relatively small enclosure leakage rate (less than 0.5 v/o per hour at 1.6 psig) can cause pressure equilibrium between a heated enclosure and its surroundings. It is concluded that only a very rapid release of heat, such as would be associated with an explosion in which several hundred kilograms of plutonium are vaporized, could lead to a significant thermal overpressure inside the Rocky Flats Plant Assembly Room.

APPENDIX III

THE MAXIMUM SAFE SOURCE STRENGTH

The maximum safe source strength may be obtained from equation (21. The Table I insoluble value for controlled areas of 4×10^{-11} $\mu\text{ci/ml}$ is assumed to be the limiting concentration. The insoluble value is chosen since plutonium dioxide has been shown to be chemically resistant to attack and can safely be regarded as insoluble (Stewart, 1963). The controlled area assumption is made since the maximum concentration occurs just ten meters from the source (cf. section C-9). There is negligible danger to off site personnel from a release originating in the Rocky Flats Plant Assembly Room since the plume will be diluted by about four orders of magnitude before reaching uncontrolled areas. The normal source area is taken as 142.3 m^2 in accord with the discussion of section D-1. Equation (21 thus gives a maximum safe free release source strength of 1.23×10^{14} Pu atoms/sec.

The use of the continuous exposure limits in the above calculations is very conservative since, according to AEC Manual Chapter 0524, these limits may be averaged over periods up to 13 consecutive weeks provided work areas are appropriately monitored and exposure histories are maintained for each individual working in such areas. The computed source strengths are thus conservative by a factor of the number of working hours per quarter divided by the number of working hours involved in the release.

ILLUSTRATIONS

Figure 1. GRAVEL GERTIE STRUCTURE

Figure 2. PARTICLE SIZE DISTRIBUTIONS

Figure 3. VALUES OF $\bar{U}X/S$ AS A FUNCTION OF DOWNWIND DISTANCE, X , (M), FOR VARIOUS WEATHER TYPES; SOURCE AT SURFACE

Figure 4-a. SOURCE STRENGTH
VARIATION WITH PARTICLE SIZE
 $M_T = 202$ kg

Figure 4-b. SOURCE STRENGTH
VARIATION WITH PARTICLE SIZE
 $M_T = 50$ kg

Figure 5-a. SOURCE STRENGTH
VERSUS EXPONENT Y

Figure 5-b. SOURCE STRENGTH
VERSUS EXPONENT Y

Figure 6. SOURCE STRENGTH VERSUS TEMPERATURE CONSTANT

Figure 7. SOURCE STRENGTH VERSUS MASS OF PLUTONIUM OXIDIZED

Figure 8-a. SOURCE STRENGTH
VERSUS LEAK PATH SIZE (0.5
v/o leakage per hour)

Figure 8-b. LEAK PATH NUMBER
VERSUS LEAK PATH SIZE (0.5
v/o leakage per hour)

Figure 9-a. SOURCE STRENGTH VARIATION WITH INERTIAL PARAMETERS

Figure 9-b. SOURCE STRENGTH VARIATION WITH INERTIAL PARAMETERS

Figure 9-c. SOURCE STRENGTH VARIATION WITH INERTIAL PARAMETERS

Figure 10. SOURCE STRENGTH VARIATION WITH LEAK PATH ROUGHNESS

Figure 11-a. SOURCE STRENGTH VARIATION WITH LEAK PATH SIZE (single leak path)

Figure 11-b. LEAKAGE RATE VARIATION WITH LEAK PATH SIZE