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STUDIES OF
THE DISTRIBUTION OF MASS RELEASED
INTO THE FFTF HOT CELL
AFTER THE DBA
INTERIM REPORT

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INTERIM REPORT

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I. SUMMARY

A. OBJECTIVE

The objective of this interim study was to determine the spatial and temporal distribution of the released mass of fuel, radionuclides, sodium, and sodium oxides within the FFTF hot cell following a 1000- and a 3000-Mw-sec DBA*, assuming various leak rates from the hot cell to the outer containment vessel.

B. PURPOSE

Knowledge of a physically realistic radiological source term is a primary need in siting analysis. In the present study, the source term depends on the manner of distribution of released mass in the FFTF hot cell following a design basis accident. Over the past several years, Atomics International has developed an experimental program and analytical techniques for studying the transport of aerosols of the kinds produced in assumed accidents. These studies have demonstrated that airborne particles which are small in size initially agglomerate and grow at rates that increase with mass concentration. The larger the particles, the faster they are removed from the atmosphere because of increase in settling velocities. With an initial high mass concentration, the removal rate is sufficiently great so that the concentration will be low in a relatively short time with little material left to leak out of the hot cell. In such circumstances, the source term for leakage to the outer containment is greatly reduced, with a consequent reduction in the leakage to the environs.

It is the purpose of this report to provide the quantitative information concerning such reductions in the source term as a function of the relevant input parameters.

C. METHOD

An empirical simplified agglomeration model that is based on experimental studies of sodium and UO_2 aerosols, and on a successful theoretical aerosol

*Design Basis Accident

agglomeration model is used. PNL⁽¹⁾ specified the following cases for study:

<u>Case</u>	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>
Available work from excursion (Mw-sec)	3000	3000	3000	1000
Na escapes to hot cell	Yes*	No**	Yes*	Yes*
Atmosphere of hot cell	N ₂	N ₂	Air	N ₂

*Free expulsion of sodium without retarding effect of either the vessel head or cell floor gamma shield;

**No release of sodium to the cell corresponding either to the absence of sodium in the reactor, or to the deflection of the sodium slug by the cover or shield.

The quantity of fuel released into the hot cell, determined from information provided by PNL, was 2 and 10% of the fuel volume in the core for the 1000- and 3000-Mw-sec accidents, respectively. An evaluation was made of the sodium mass released into the hot cell following the DBA based on the fuel volume vaporized. The relevant information was programmed into the empirical model with computation of the following data: airborne mass concentration, accumulated masses on floor and wall, and mass leaking from the hot cell, each as a function of time. The mass leakage depended on the choices of assumed leakage rates which were 10, 50 and 100% of the volume per day.

A fraction of the fission product inventory produced after 200 days at 400-Mw power was assumed to be released and scavenged by the fuel* and sodium*, or by the fuel alone. The released fission products, except for the rare gases, have mass distributions which are partitioned between walls and floors in the same proportion as the masses of fuel and sodium. The fraction of the total mass which leaks from the hot cell is thus representative of the fraction of any material present; thus the amount of leakage of a given material is the product of the leakage fraction and the released quantity of that material.

*In the form of an aerosol.

D. CONCLUSIONS

The spatial and temporal behavior of the material released into the hot cell of the FFTF following the design basis accident has been determined. The basis of the calculations is the information derived from experiments performed at Atomics International with a Laboratory Test Chamber (LTC). A simplified Engineering Mass Model (EMM) was used for released mass concentrations up to 200 $\mu\text{gm/cc}$. The fraction of the released mass which leaks increases exponentially with time; and, at infinite time ($t = \infty$) is directly proportional to the assumed leakage rate from the hot cell to the outer containment vessel, and is an inverse function of both the released mass and the effective density of the material. At $t = \infty$ the leakage fraction is given by:

$$f_L(\infty) = \frac{5.0 \gamma}{(\rho_R d)} 0.8,$$

with

$f_L(\infty)$ = leakage fraction,

5 = a constant for hot cell geometry,

ρ_R = initial mass concentration in the hot cell ($\mu\text{gm/cc}$),

d = the effective density of the material (gm/cc), and

γ = the leakage rate (1/day).

If fallout were not present as a removal mechanism, all the material would eventually leak from the hot cell. Thus, $f_L(\infty)$ is the reduction fraction attributable to agglomeration combined with fallout. An example is given for the case of 72 kgm of PuO_2 released in the 3000-Mw-sec accident, and a leakage rate of 0.1 per day; i. e., in the cases considered, $f_L(\infty)$ ranges from 0.005 to 0.01, corresponding to the release of 350 to 720 gm of PuO_2 to the outer containment vessel.

It is estimated that the uncertainty in the computed leakage fractions does not exceed a factor of 1.5. However, a number of conservative assumptions were made in the calculations which may counterbalance the uncertainty factor. These assumptions include the use of constant leakage rates. However it is likely that particulates passing through small cracks and pores will deposit and eventually clog such pathways, corresponding to leakage rates which

diminish with time. Additionally, credit may be taken for PuO_2 fallout in the outer containment vessel, further reducing the source term for computing the downwind dose in a radiological analysis.

There are several conservative assumptions inherent in this study, which are recommended for further evaluation. In order to make an even more convincing argument about the reduction of reactor hazards due to agglomeration, the following additional topics are suggested for study.

- 1) It is assumed that the particulates which are formed will leak through the hot cell leak paths in the same manner as a gas. Estimates of the reduction of particulate leakage due to impaction and fallout in various assumed leak paths should be made.
- 2) It is assumed that the mass which has leaked into the outer containment does not settle, but remains airborne for long time periods and is therefore available for leakage to the site environment. Estimates of the reduction in concentration due to settling in the outer containment can be made from the knowledge of the particle sizes which have leaked from the hot cell. The particle sizes in the hot cell were computed in the current study.
- 3) All calculations assume that the leak rate γ is a constant throughout the time of interest. The leak rate from the hot cell is a function of temperature and pressure. The variations of these parameters with time should be computed from a thermal analysis of the energy released to the hot cell and the resulting decrease of temperature and pressure due to heat losses.
- 4) In evaluating the material which leaks out of the outer containment one should remember that the design pressure is not reached instantaneously but that the pressure rises as gas escapes from the hot cell. When the leak fraction from the hot cell is known as a function time (Recommendation No. 3), the pressure rise in the secondary containment can be used to determine the leak fraction to the environment. The leak rate at 2 hours is considerably below that of the design pressure and effectively reduces the mass escaping to the environs.

II. INTRODUCTION

Current practice in the evaluation and design of safeguards for reactors takes almost no credit for diminution of the potential radiological source term as demonstrated by numerous experiments and analytical studies on aerosol agglomeration.

Studies are currently being conducted at Atomics International under the AEC Nuclear Safety Program* to develop experimental information and analytical methods which characterize the release and transport of effluents generated during major Liquid Metal Fast Breeder Reactor accidents. The information developed under this program and others has provided sufficient background and evidence to support a more realistic assessment of the radiological source term for the most severe fast reactor accidents.

The study described in this report represents the correlation and application of experimental and analytical modeling data to the evaluation of the fate of fuel, sodium, and radionuclides released as a result of the postulated design basis accident (DBA) for the FFTF. By using the existing knowledge of the behavior of aerosols, it is shown that the potential hazards of the DBA are greatly reduced. This reduction occurs because of two mechanisms: (1) the use of double containment and the resultant low leak rate from the outer containment at early times after the DBA and (2) the predominant reduction of the source term produced in the inner containment system (FFTF hot cell) by the agglomeration and fallout of the materials released during the accident.

In order to justify the hypothesis used in this analysis, a brief description of the state-of-the-art knowledge about aerosols, agglomeration, and fission product deposition on aerosols is presented. A correlation between laboratory experiments and modeling techniques is shown to be possible through the use of rigorous physical principles. Next, the derivation of an empirical relationship for characterizing aerosol behavior is shown to be consistent with results from laboratory tests. The development of an engineering mass model (EMM) programmed for solution on a digital computer is presented and its general agreement with laboratory data is substantiated.

*Sponsored by AEC-DRDT Environmental and Sanitary Engineering Branch.

This analytical tool was then utilized for a parametric study of the spatial and temporal distribution of the materials released to the FFTF hot cell following the DBA, for various specified conditions. Finally, the results of these computations are summarized and discussed in terms of their significance and application to safeguards evaluation.

III. DESIGN BASIS ACCIDENT SOURCE TERMS

The design basis accident for the FFTF reactor as postulated by PNL for this study is an excursion with a total energy release in the range 1000 to 3000 Mw-sec which results in vaporization of a fraction of the core. The resultant fuel vapor volume expands out of the core releasing fuel (PuO_2 and UO_2), fission products, and sodium coolant to the hot cell environment. Free expulsion of sodium without any retarding effect of the vessel head or the cell floor gamma shield is assumed. In the present analysis, it is also assumed as suggested by PNL that the fraction of the fuel which is vaporized is 2 and 10%, for the 1000 and 3000 Mw-sec excursions respectively. The fuel mass associated with these fractions is assumed to be released as vapor into the hot cell inner containment. A mass of sodium is expelled concurrently from the pool above the core; and, for a first approximation, it is assumed to represent a slug of sodium, 15 ft. high with a diameter equal to that of the vaporized fuel volume.

The resultant mixtures of fuel, sodium, sodium oxide, fission products, and other reactor materials are airborne and thus represent the potential source term for the evaluation of the radiological dose at the reactor site. The detailed computation of the actual quantity of these materials is shown in Section VIII. The ultimate source term which directly controls the site selection is the quantity which leaks from the outer containment. The present study is concerned with the development of a suitable model which allows one to compute the accumulation of material in the outer containment. The actual mass which escapes to the $4 \times 10^6 \text{ ft}^3$ outer containment volume from the hot cell is a function of the leak rate from the FFTF hot cell and the agglomeration, growth, and settling characteristics of the aerosol.

IV. REVIEW OF AEROSOL CHARACTERISTICS

A. COAGULATION (AGGLOMERATION)

The property of continuous and spontaneous coagulation is a striking characteristic of aerosols. The aerosol particles, of whatever substance they are composed, coalesce or stick together when they come into contact, and the process goes on continuously so that all aerosol particles become larger and finally settle. The first clear evidence of coagulation in smoke was obtained by Tolman (1919)⁽²⁾. He found that while for any one smoke the mass concentration in a confined volume of smoke decreased linearly with time, the intensity of light scattered by the smoke diminished more rapidly and gave a curved graph. By independent experiments, it was found possible to correlate the intensity of the scattered light with the mass concentration and with the average size of particle; it then became evident that the decrease in scattered light intensity can be explained by collision and coagulation of the particles in the aerosol. These experiments however, gave no information about the number of particles present at any instant or their size. The true nature of the coagulation process in smokes was elucidated in the researches of Whytlaw-Gray (1932) and his co-workers⁽³⁾, who first developed reliable methods of counting the particles in smoke clouds and then used the information to determine the variation of the number of particles with time; i. e., the form of the coagulation curve. Data they obtained show that the coagulation of very many smokes follows a simple law. If n is the number of particles per cm^3 at any time t , and n_0 is the number present when the smoke is formed, then:

$$\frac{1}{n} - \frac{1}{n_0} = Kt, \dots \quad \dots (1)$$

where K is a constant. This, if the reciprocal of the number of particles present in a given volume of smoke is plotted against the corresponding time, a straight line is obtained in nearly all cases. If $1/n = P$ and $1/n_0 = P_0$, we have

$$P = P_0 + Kt, \dots \quad \dots (2)$$

where P has a definite physical meaning and represents the average volume occupied by a particle in the smoke, and the slope of the curve is the coagulation constant K . If the particles are assumed to be monosize, the particle volume P equals $4\pi/3 d r^3 / (M/V)$ where d is the particle density and M/V is the mass concentration of the particles; therefore, the particle radius r is related to K and t :

$$\frac{4\pi d (r^3 - r_o^3)}{3 \left(\frac{M}{V}\right)} = Kt \quad \dots (3)$$

At a fixed time t , and when $r_o \ll r$,

$$r^3 \approx \frac{3}{4\pi d} \left(\frac{M}{V}\right) Kt \quad \dots (4)$$

$$\text{or } r \approx \text{constant} \left(\frac{M}{V}\right)^{0.33} .$$

Figure 1 is a graph of Equation 4, which shows the dependence of r in microns on time and the mass concentration (M/V). The value used for K was 3×10^{-10} cm³/p-sec, which is the agglomeration constant consistent with the Brownian motion of large particles.

If an additional mechanism such as gravitation increases the magnitude of K , the time t required to reach a certain size will decrease.

In a fixed geometry, the growth of the particles are limited since, after they reached a certain size, their settling rate becomes so large that they are removed from the chamber; this is in contrast to the simple Whytlaw Gray relation where the particles grow forever. The time required for particles to reach a given agglomeration size varies with the mass density; therefore, if both settling and agglomeration occur, a maximum agglomerated particle size should be produced in a given size chamber.

Equation (1) is of the same form as that established by Rutherford (1897)⁽⁴⁾ for the rate of combination of small ions and that known to be valid for the combination of large atmospheric ions⁽⁵⁾ and for the rate of disappearance of

Figure 1. Particle Growth vs Time and Mass Concentration
(Due to Brownian diffusion)

nuclei;⁽⁶⁾ naturally on account of the smaller mobilities, K is smaller for smoke particles than for ions. Oppositely⁽⁷⁾ charged ions recombine at a rate 10^4 times that of particles of about 1μ .

From Equation (1), by differentiation, one obtains the rate at which the particles disappear by coagulation. This depends only on the square of the number of particles present and the constant K ; i. e. ,

$$-\frac{dn}{dt} = Kn^2 \quad \dots (5)$$

Thus, coagulation is very rapid in the early stages when the number concentration is high. A comparison of the value of the coagulation constant K , obtained experimentally, for many different aerosols in air under normal conditions of temperature and pressure, indicates that although K varies from smoke to smoke, the coagulation process does not depend markedly on the nature of the aerosol.⁽⁸⁾

The rapidity of coagulation in particulate clouds of high number concentration is better appreciated when the time taken to halve the number of particles is calculated. This is found from Equation (1) by use of $n = n_0/2$ hence $t = 1/n_0 K$. By taking the value of K for a smoke as $0.51 \times 10^{-9} \text{ cm}^3/\text{sec}$, we find for a system of 10^{11} particles per cm^3 , that $t = 0.02 \text{ sec}$; for 10^6 per cm^3 , 33 min; and for 10^5 per cm^3 , 5.5 hours. Therefore, when the number concentration is high, coagulation is so rapid that the number per cm^3 present after a few minutes is practically independent of the initial number.

The agglomeration constant K , is not the same value for all particles sizes or under all physical conditions. It is larger for heterogeneous distribution than for monosize distribution. The coagulation rate is also increased by air motion (turbulence) and by differential settling. In the latter case, heterogeneous aerosols coagulate even more rapidly when large particles fall past the smaller ones and cause further collisions.

B. CHARGE EFFECTS

Due to the radiation that is present during the postulated accident, the effect of ion pair production and recombination on aerosol behavior is of concern. It is reasonable to expect that equal numbers of oppositely charged ions and electrons are produced and that the affinity of the two charge polarities for aerosol particles is not preferential. The aerosol particles will therefore become to some degree bipolarly charged.

Zebel⁽⁹⁾ has shown that the agglomeration rate will depend on the degree of charging or specifically on the ratio of the number of elementary charges (e) carried by the colliding particles, to the size of the particles. His formulation gives for the ratio (z), of the agglomeration rate for a charged aerosol, to the rate for an aerosol under the influence of Brownian motion alone, the value

$$z = \frac{y}{e^y - 1} \quad \dots (6)$$

where:

$$y = \frac{\frac{nme^2}{kT}}{r_n + r_m}$$

$r_n + r_m$ = contact distance of the colliding particles,

kT = thermal energy, and

n, m = numbers of elementary charges on each of the two particles (n, m are positive or negative according to the type of charge involved; for the case under discussion, n and m are respectively positive and negative; therefore, the product must be negative.

Thus, for the present situation, y is a negative quantity.

A sketch of Equation (6) is shown in Figure 2. ⁽¹⁰⁾ From this information, the agglomeration rate for a weakly charged bipolar aerosol, $|y| \ll 1$, is only

Figure 2. The Function Z vs Y ($y = 0$ corresponds to an uncharged particle)

slightly greater than that for an uncharged aerosol. For a strongly charged bipolar aerosol, y is a large negative number; in this case agglomeration takes place at a much greater rate and consequently settling of the particles will be substantially accelerated.

Inasmuch as a reasonable postulation of circumstances which would lead to a monopolar aerosol has not been established, inhibition of agglomeration is not expected.

The coagulation constant of nonradioactive and radioactive metallic aerosols produced by an exploding wire technique has been determined experimentally by Rosinski.⁽¹⁰⁾ The temperature of the exploding wire was estimated to be 7000 to 8000°K. Rosinski found that in the early stages of agglomeration, the coagulation constant of radioactive gold aerosols (2 to 3.5 C_i /gm) was approximately 20 times the mean value of slightly radioactive aerosols (50 to 900 mC_i /g). He assumes that coagulation is enhanced by the presence of highly ionized gas.

In the present analysis, $\sim 3 \times 10^7 C_i$ of activity is present in the hot cell volume after the DBA. The production rate of ion pairs, q , is $\sim 3 \times 10^{12}$ ion pairs/cc-sec. The recombination coefficient α is $\sim 2 \times 10^{-6}$ cm^3/sec per particle.⁽¹¹⁾ The rate at which the ion concentration varies with time is:

$$\frac{dn}{dt} = q - \alpha n^2$$

and at equilibrium, $\alpha n^2 = q$. Thus the equilibrium concentration of uncombined ions is $\sim \sqrt{1.5} \times 10^9$ ion pairs per cm^3 . These ions will readily attach to any neutral or charged particles which may be present as a result of the reactor accident.

The mass median initial radius of the sodium oxide and vaporized UO_2 is ~ 0.5 and 0.3 microns, respectively; therefore, the number density of particles, N , for a given mass concentration, ρ_R , is

$$N = \frac{10^{-6} \rho_R}{\frac{4\pi}{3} r_o^3 d}$$

where:

ρ_R = the mass concentration in $\mu\text{gm/cc}$,

d = the particle density in gm/cc , and

r_0 = initial mass median particle radius in cm ($1\mu = 10^{-4} \text{ cm}$).

If ρ_R is assumed to be $25 \mu\text{gm/cc}$ of either U_3O_8 (the product produced when UO_2 is vaporized in air) or Na_2O , $N = 3 \times 10^7$ particles/cc, assuming all particle radii to be the same as the mass median radius. This instantaneously produced concentration is in the presence of 10^9 ion pairs per cm^3 . N , the particle number density, decreases with time due to agglomeration, but the ion pair density varies slowly, decreasing only with radioactive decay. The rate of decrease of the particles and ions are represented by KN^2 and λn , respectively, where λ is the radioactive decay rate and K is the agglomeration rate.

If
$$\lambda = \frac{0.69}{T_{1/2}(\text{sec})} = \frac{0.69}{0.86} \times 10^5 \text{ sec},$$

and
$$K = 3 \times 10^{-10} \text{ cm}^3/\rho\text{-sec}; \text{ then,}$$

$$KN^2 = 3 \times 10^{-10} \times 9 \times 10^{14} = 27 \times 10^4 \rho/\text{cc-sec},$$

and
$$\lambda n = 0.8 \times 10^{-5} \times 10^9 = 0.8 \times 10^4 \rho/\text{cc-sec}.$$

Thus N decreases much faster than n , if the particles are neutral. If the N particles are positively charged, these particles would be in competition with the negative ions produced by ionization and would rapidly become neutral. One may therefore conclude that there will always be an excess of ion pairs, which in turn can only produce a bipolarly charged system of particles. As was shown earlier, such a system produces more rapid agglomeration than an uncharged particle system.

V. FISSION PRODUCT SCAVENGING

Following a nuclear reactor accident, the presence of airborne fission product nuclides in particulate form has always been considered a distinct possibility. The design basis accident postulated for the FFTF reactor involves the vaporization of a fraction of the core with the simultaneous release of at least the same fraction of the fission product inventory.

In the present evaluation, it will be shown that sufficient evidence is available from experimental and modeling data to enable determination of a reasonable representation of the particle size distribution or its mass median radius which forms following the release of large masses of material into a relatively leak-tight enclosure. The effects of agglomeration and the resultant increase in particle size and settling rate is based on a thorough literature search, and on the aerosol studies in progress at Atomics International. Fission product scavenging by the larger particulates is also well supported by numerous investigators.⁽¹²⁾

A review of the literature on fission product release studies conducted under the AEC Nuclear Safety Program reveals that the predominant effort has been in the area of gaining an understanding of the behavior of iodine in molecular, acidic, or organic form.⁽¹³⁾ In these studies, various quantities of iodine have been released into vessels of different sizes and the rate of disappearance of iodine has been measured. In other tests, gross fission products including I_2 have been released from molten UO_2 and/or uranium.^(14,15) All of these experiments are based upon accidents postulated for the pressurized water reactor; and it is assumed that the fuel melts, and only fission products are released. Under these conditions, the mass of fission products released to the containment vessel is very small; thus, for large volumes, the airborne mass concentration is also very low. If the fuel temperature does not exceed the melting point, the concentration of vaporized fuel is also very low. When only I_2 release is simulated, there is essentially no mass upon which the I_2 can plate, except the walls of the test vessel.

In essentially all of these studies, even at these low concentrations, many of the fission products were found to be in the form of particulates. However,

the emphasis was almost always placed on the unagglomerated fraction and the fate of the molecular I_2 or CH_3I (methyl iodide).^(13,15,16,17,18)

Studies which strongly supported the agglomeration or adsorption of iodine were designed to show that desorption of the iodine would occur even though it required temperatures well in excess of the sublimation temperature of iodine, in order to demonstrate desorption.⁽¹²⁾ Of course, if the iodine reacted with the particulate material, desorption could not be demonstrated.

In the study of the agglomeration of natural decay products of Rn on aerosols, the experiments were carried out at very low concentrations of the aerosols, and thus the agglomeration of the material to which the radionuclide becomes attached was not studied.^(19, 20, 21, 22, 23) However, it was clearly demonstrated that the natural and artificial radioactive contaminants which are in molecular or atomic states become attached to the fine solid or liquid particles which are always present.

Another series of investigations are being conducted on the ability of water and hydrazine sprays to absorb and react with radioactive compounds of iodine and thus effectively remove these radioactive compounds from the reactor vessel before leakage to the environs can occur.^(24, 25, 26, 27) The theories for the iodine scavenging ability of water sprays with large particle sizes (100 to 300 microns in mass median diameter) is almost identical to that proposed for explaining the attachment of natural or artificial radionuclides on aerosols.^(28, 29, 30) In essence, it is the rapid diffusion of the small particles (i.e., iodine or other fission products) across a boundary to the surface of the almost stationary large particle which removes the radioactive particle. Of course, the rapid fallout of the large spray droplets insures that the source term for dispersion of the iodine downwind is negligible. It is this same mechanism of the fallout of a large particle which reduces the source of PuO_2 and scavenged fission products in the present study.

On the basis of the above discussion, it is assumed for the present study that the fission product mass is scavenged by the released sodium and fuel.

Thus the fraction of the total mass which leaks from the hot cell represents the fraction of the total activity which (corrected for decay) leaks. Only the noble gases are unagglomerated and are thus completely available for leakage from the hot cell.

VI. MONOSIZE AGGLOMERATION MODEL DEVELOPMENT

A. SUMMARY OF AI AEROSOL EXPERIMENTS^(31, 32, 33, 34, 35, 36)

The characteristics of sodium and UO_2 aerosols have been investigated in a laboratory test chamber (LTC) 40-ft³ volume, 6-ft high by burning a pool of liquid sodium in various oxygen environments, or by arc-vaporization of UO_2 in stainless steel containers. Following the release of material into the chamber, sequential samplers automatically collect materials on the walls, floor, and in the air. Manually taken samples of the air concentration are obtained with round jet cascade impactors and electrostatic precipitators for particle size distribution measurements. Cumulative samples are obtained on the walls and floor, and a mass balance between the sum of the material on the wall and floor and that remaining airborne represents the total mass released. After the aerosols formation deposition samples collected at the floor and at any distance from the floor have shown equal amounts of mass and thus demonstrate "uniform concentration conditions". In these tests, the floor and wall deposition rates of the released materials are measured as a function of time. Graphs of the cumulative deposits of material allow one to determine (a) the total amount deposited and (b) the fraction of the material which is deposited during a specific period of time.

There are four observable phenomena which conveniently characterize an experiment. All of these show a dependence on the released mass concentration, $M_R/V = \rho_R$ (μ_g/cc). These relations, the method of their determination, and their dependence on ρ_R and other input functions are discussed below.

- 1) $M_F/M_R(\infty)$ is the fraction of the released mass which is ultimately deposited on the floor. It is the ratio of the mass deposited on the floor divided by the mass on both the floor and wall after all of the airborne mass has been removed. The value of $M_F/M_R(\infty)$ increases as ρ_R increases.
- 2) ρ_{max} is the maximum airborne mass concentration in $\mu\text{gm}/\text{cc}$. ρ_{max} occurs at the end of τ , the burning period. The ratio ρ_{max}/ρ_R decreases as ρ_R increases and as τ increases.

- 3) T_{50} is the time for 1/2 of the mass to be deposited on the floor after burning has ceased. As ρ_R increases T_{50} decreases.
- 4) $r_{\max}$ is the maximum mean particle radius that the initial mass distribution grows to during the life of the aerosol. There are three methods used for characterizing $r_{\max}$. These are respectively (a) r_m , from the floor deposition rate (b) r_{50} , from the floor deposition half time, T_{50} , and (c) r_a , from measurements of the particle size distribution of the mass concentration determined by using cascade impactors, see Section VII.

In all cases, the measured $r_{\max}$ has a relation to ρ_R of the form $r_{\max} = a(\rho_R)^b$. The coefficient, a , varies from 0.8 to 1.0; and b , varies from 0.4 to 0.55. The relation of $r_{\max}$ to ρ_R for arc-vaporized UO_2 in air also fits the above range of values for a and b , even though the vaporization time is much less than the burning times for sodium.

The conclusions derived from the LTC experiments in which ρ_R was varied from 2-60 $\mu\text{gm/cc}$ follow.

- 1) As more aerosol is released into the chamber, the time for clearing the air decreases. The particle size distribution increases to larger maximum sizes as the mass of material released is increased, regardless of the shape of the aggregate (chains or spheres). The relation with the maximum concentration has been found to be approximately $r_{\max} = \rho_R^{0.5}$, where ρ_R is in $\mu\text{gm/cc}$ and $r_{\max}$ is the maximum mass mean radius in microns.
- 2) Wall plating rates indicate that an increase in the rate of deposition occurs with increase in the airborne concentration which is associated with higher temperature gradients.
- 3) The initial size distribution of the released sodium oxide is log normal, with an initial mass median radius, r_o , of 0.5μ and a geometric standard deviation of 2.33. The initial mass median radius, r_o , of the $U_3O_8 - Fe_3O_4$ aerosol is 0.3μ with a geometric standard deviation of 2. In all of the experiments, the radius changes in time to a value $r_{\max}$. The time for $r_{\max}$ to occur decreases as ρ_R increases and τ decreases.

- 4) Mixtures of aerosols produced by arc-vaporizing UO_2 -stainless steel in air and by burning liquid sodium simultaneously were also studied. Both material agglomerated together to form a complex branched aggregate which settled to the floor at a rate proportional to the mass concentration of the combination.

Work performed at Brookhaven National Laboratory⁽³⁷⁾ indicates that aerosols from vaporized PuO_2 behave identically to those of UO_2 and when vaporized together, they are indistinguishable as aggregates under high magnification with an electron microscope. BNL experiments with vaporized Na and PuO_2 show that the materials combine to form a single aggregate with a concentration that diminishes in proportion to the effective density of the aerosol.

B. DESCRIPTION OF THE ENGINEERING MASS MODEL (EMM)

The EMM is constructed to account for the transport of airborne particulates, including floor fallout, wall plating, and removal from a chamber by the mechanisms of leakage and/or ventilation. The EMM is an empirical representation and is based on:

- 1) experimental data on U_3O_8 and sodium oxide aerosols obtained in the LTC and
- 2) a heterogeneous aerosol agglomeration code (HAA)⁽³⁸⁻³⁹⁾ that has been used successfully to represent mass distribution data in the LTC experiments for mass concentrations up to $\sim 10 \mu\text{gm/cc}$. An example of the typical agreement obtained between experimental data (see squares) and code produced data (see 1's) is illustrated in Figure 3a, b, c for test no. 5 ($\rho_R = 2.5 \mu\text{gm/cc}$), $\tau = 5.5 \text{ min}$. The ordinates are suspended component, settled component, and average radius in microns, each as a function of time.

In this EEM model, particles will "grow" in size, fallout will increase, etc. The term used to represent fallout is the expression based on the Stokes velocity for a falling particle in a gravitational field, and is the same as the term employed in the HAA code. Similarly the wall plating term is taken from this agglomeration code. It is an empirical expression describing diffusion

Figure 3a. Comparison of HAA Code and Experimental Data

Figure 3b. Comparison of HAA Code and Experimental Data

Figure 3c. Comparison of HAA Code and Experimental Data

to the walls. There are two major simplifications used in the EMM: first, a monosize model is employed in which a single radius, r , represents a mean particle size; second, the terms in the HAA code, used in the complete integro-differential equation, which represents the growth of particles, are replaced by the simplifying assumption that the mean size, r , is a stated function of the instantaneous airborne mass concentration, ρ , at any time; i. e. ,

$$r = r_0 + a\rho^b .$$

The initial task in formulating and testing the EMM, was to determine whether or not a set of values for r_0 , a , and b exist that will adequately represent the accumulated data of the LTC experiments. The only adjustable parameter in the model is the "plating-distance", Δ , appearing in the wall-plating term of the HAA code. The testing of the EMM is accomplished by comparing model computations of the functional dependence on ρ_R of the four observable factors: $M_F/M_R(\infty)$, T_{50} , ρ_{max} , and r_{max} . These were defined in an earlier part of this section.

The extent to which the EMM formulation is in agreement with the experimental data is a measure of the model's applicability, and will provide an estimate of its accuracy in the analyses of the consequences of the FFTF DBA.

C. ENGINEERING MASS MODEL EQUATIONS

The basic differential equation describing the mass distribution in a given volume is given by:

$$\frac{d\rho}{dt} = S_M - \alpha\rho - \beta\rho - \gamma\rho , \quad . . . (7)$$

where:

α, β, γ , = removal rates from the airborne system due, respectively, to floor fallout, wall-deposition and leakage in sec^{-1} ,

ρ = airborne mass concentration, $\mu\text{gm/cc}$,

$$S_M = \frac{M_R}{V\tau}, \text{ } \mu\text{gm/cc-sec, and}$$

$$V = \text{volume, cm}^3 \text{ (for LTC } V = 1.13 \times 10^6 \text{ cc).}$$

The removal rate, α , is related to the Stokes expression for particle velocity and to geometrical factors

$$\alpha = v_s \frac{A_F}{V} \text{ with } v_s = \frac{2}{9} \frac{gd r^2}{\eta} \quad \dots(8)$$

where:

$$A_F = \text{floor area, cm}^2 \text{ (} 6.26 \times 10^3 \text{ cm}^2 \text{ for LTC),}$$

$$v_s = \text{Stokes velocity in cm/sec, for a particle with radius } r,$$

$$g = \text{gravitational constant, cm/sec}^2,$$

$$d = \text{particle density, g/cc, and}$$

$$\eta = \text{air viscosity, gm/cm-sec. (at } 300^\circ\text{F)}$$

With r expressed in μ , v_s in cm/sec, and d for sodium oxide as 2.27 gm/cc, the expression becomes:

$$v_s = 2.71 \times 10^{-2} r^2 .$$

If particle sizes are much smaller than 1μ , a Cunningham correction factor is to be added. Thus, $\alpha = 1.50 \times 10^{-4} r^2$; (r in μ and α in sec^{-1}).

The removal rate β , is related to the geometrical constants and the diffusion constant as follows:

$$\beta = v_w \frac{A_w}{V} , \quad \dots(9)$$

where:

$$v_w = \frac{D}{\Delta},$$

v_w = plating velocity at the walls, cm/sec,

D = diffusion coefficient of a particle with radius r , cm^2/sec , and

Δ = plating distance constant, microns.

A_w = wall area cm^2 (for LTC $A_w = 1.035 \times 10^5 \text{ cm}^2$); with
 $D = 4/3 kT/\eta l/r$, one obtains $v_w = 1.18 \times 10^3 (1/\Delta \cdot r)$ and
 $\beta = 1.08 \times 10^{-4} (1/\Delta \cdot r)$ for the LTC.

The use of Expression (9) for β and v_w does not imply that a simple diffusion model represents the plating action on the wall. Rather, Expression (9) is to be regarded as an empirical formulation which represents the data reasonably well, and uses " Δ " as an adjustable parameter, which depends largely on the released density, ρ_R .

The leakage rate, γ , is equal to zero in the case of the LTC. When applicable, γ represents the fractional leak rate out of the container due to leakage and/or ventilation. It may be a constant, unlike α and β , which depends on the magnitude of the mean radius.

The final form of Equation (7) is now:

$$\frac{d\rho}{dt} = S_M - 1.50 \times 10^{-4} r^2 \cdot \rho - 1.08 \times 10^{-4} (1/\Delta \cdot r) \rho \quad \dots (10)$$

The source term

$$S_M = M_R/V \cdot \tau = \rho_R/\tau \text{ for } 0 \leq t \leq \tau \text{ and } S_M = 0 \text{ for } t > \tau.$$

As stated earlier, the effect of agglomeration is represented by using a functional dependence between r and ρ ;

$$r = r_0 + a\rho^b \quad \dots (11)$$

The adjustable constants r_o , a , b , and Δ are chosen so as to give the best "fit" to the previously described sets of data obtained from the LTC tests.

D. SELECTION OF EMM CONSTANTS FOR AGGLOMERATION EFFECTS

The values for the three constants r_o , a , and b , in the EMM were selected so that simultaneous representation was possible for four independent sets of data. The range of values of ρ_R in the LTC series was 2 to 60 $\mu\text{gm/cc}$. The observed mass distributions are affected by the "geometry" in the case of the LTC experiments because of the relatively large wall area. In fact, for the lower ρ_R values, the walls receive about 40% of the total mass available. Thus we must properly account for these effects in the LTC to be sure that the removal terms are correct when scaling to FFTF where wall plating is not important.

The choice of the model used to describe agglomeration effects that represented all the data is

$$r = 0.2 + 0.7 \rho^{0.5} .$$

The extent of the "fit" can be summarized briefly

1. Floor Deposition, $M_F(\infty)/M_R$

The EMM calculations fit the observed data over the range of ρ_R values with a set of " Δ " values that were consistent with those used in the HAA code.

2. Floor Half Time, T_{50}

The computed values fit the floor half time data over the range of ρ_R values, with a set of " Δ " values consistent with the ones for floor deposition.

3. Maximum Airborne Mass Concentration, $\rho_{\max}$

The computed values fit the measured $\rho_{\max}$ (values range over a factor of 10) to within 15%.

4. Maximum Mean Particle Radius, $r_{\max}$

The computed $r_{\max}$ values follow the correct slope (as a function of ρ_R) as compared with observed values. The latter constitute a "band" rather than a line, since three independent " $r_{\max}$ " values are measured in the experiments, as explained previously. The computed $r_{\max}$ values are within 20% of an overall "mean" experimental line. On the basis of the comparisons made, the model $r = 0.2 + 0.7 \rho^{0.5}$ used in conjunction with Equation 10, may be expected to yield results valid to within 20% for the FFTF calculations.

5. Temperature Effects

The settling and wall plating velocities are both functions of temperature. In the LTC studies, the average temperature in the test cell changes with time. The mean temperature during the early life of the aerosol is used in the comparisons which were made. A temperature of 300°F was used for the FFTF computations.

VII. FFTF HOT CELL MODEL

The mass of fuel and sodium which may be released into the FFTF hot cell following the DBA is large compared to that used in the LTC tests. However, it has been shown that it is the released mass concentration, ρ_R , which determines the governing aerosol characteristic, the mean particle radius by mass. As mentioned in Section III, the mass of fuel vaporized and available for release has been specified by PNL as being 2 and 10% of that in the core.

The detailed processes associated with the expulsion of some fraction of the sodium pool above the core following the DBA are complex in nature, involving appropriate treatment of energy partitioning, fuel vapor thermodynamics, and hydrodynamics of a pressure source expanding within and against a liquid column. The detailed analysis of this problem with all of the above considerations was beyond the scope of the present study. However, the results from such an analysis would still contain uncertainties owing to the assumptions required in those areas where a completely rigorous treatment can not be formulated.

For a preliminary evaluation of this problem, it is assumed that the amount of sodium which could be expelled, from the pool (above the core) is directly related to the effective cross-sectional area of the vaporized fuel volume acting as a piston (and pressure source) against the sodium column. The initial fuel vapor/volume (at the termination of the DBA) could expand out to the reactor vessel wall expelling sodium in a quantity approximating that of the entire pool of sodium, if the retarding effects of the vessel head and the cell floor gamma shield are neglected. However, it is possible for the initial fuel vapor volume to vent through the sodium pool to the hot cell after ejecting a slug of sodium considerably smaller in diameter than the vessel diameter, and hence involving a much smaller volume of sodium than would otherwise be the case.

Thus, the concept of expansion of the fuel vapor leading to expulsion of the entire sodium pool is in competition with the venting mechanism considered and the corresponding smaller fraction of the sodium leaving the pool. A preliminary comparison of the ejection time for a slug of sodium, 15-ft. high and having a diameter equivalent to that of the vaporized fuel volume, with the time for

expansion of the same volume out to the reactor vessel wall indicates that slug ejection (and venting) could occur somewhat earlier. The time for ejection of this slug is of the same order of magnitude as the time required for the radius of the fuel vapor volume to increase to several times its original value. For the purpose of this study, early slug ejection and venting was assumed, and the mass of the sodium slug ejected was varied to simulate variations in initial fuel vapor volume. One case involving the entire sodium pool was also selected as an upper limit.

The simplified model adopted for estimating the mass of sodium expelled from the core, assumes that the fraction, F , of the 3151 kg of fuel which is released occupies a sphere whose volume is

$$V_{fv} = V_f \cdot F;$$

$$V_{fv} = \frac{3151 \text{ Kg} \cdot F}{\text{density of fuel}} = \frac{\pi}{6} D^3$$

D the diameter of this sphere equals the diameter of a cylinder whose height is the 15 ft of sodium in the pool above the FFTF reactor core; thus, the volume of sodium which is expelled, unrestricted, into the hot cell is $\pi D^2/4 \times 15 \text{ ft}$. The airborne concentration of both fuel and sodium is assumed to occur uniformly and instantaneously into the hot cell volume.

The resultant $M_{R/V} = \rho_R (\mu\text{gm/cc})$ is the parameter which determines the maximum mass median particle radius $r (\mu)$, which in turn controls the ultimate fate of the airborne mass.

Estimates of the mass of sodium and fuel which are released based on the above analysis are shown in Table 1.

As shown in the EMM development (Section VI) the dimensions of the enclosure control the removal constants α and β (1/sec). Similarly, it is the ratio of γ , the leak rate, to $\alpha + \beta$ which determines the fraction of the released material which leaks to the outer containment vessel. Neither α nor β is a constant, since both are functions of r and therefore of ρ which varies with time.

TABLE 1
INPUT PARAMETERS FOR ESTIMATING EFFECTS OF ACCIDENTS

Parameter	Specified Case No.				New Case No.		
	A	B	C	D	E	F	G
Energy of the Excursion (Mw/sec)	3000	3000	3000	1000	3000	3000	3000
Hot Cell Atmosphere	N ₂ [*]	N ₂	Air	N ₂	N ₂	Air	Air
Na Escape	Yes	No	Yes	Yes	Yes	Yes	Yes
Fuel Vaporized (%)	10	10	10	2	10	10	10
Fuel (kg)	315	315	315	53	315	315	315
PuO ₂ Vaporized (kg)	72	72	72	14.4	72	72	72
Na Released (lb)	970	-	970	330	2000	3000	40,000
Na Released (kg)	440	-	440	150	910	1380	18,000
Na ₂ O Airborne	600	-	600	210	1225	1870	3000†
F. P. Released§ (kg)	17.4	17.4	17.4	4.4	17.4	17.4	17.4
M _R , Total Mass Airborne (kg)	932	332	932	267	1557	2200	3100
f _p (Fraction PuO ₂ of M _R)	0.0785	0.22	0.0785	0.054	0.046	0.033	0.0232
f _{FP} (Fraction of FP of M _R)	0.019	0.052	0.019	0.0165	0.011	0.008	0.0057
Mass Concentration (M _R /V=ρ _R)							
Total (μgm/cc)	74.5	26.6	74.5	21.4	125	176	240
Density, d _{eff} (gm/cc)	3	8.0	3	2.8	2.65	2.5	2.3

†18,000 kg released; 11,000 kg burned in 2 hr; 2,200 kg released in 2 hr = 3000 kg Na₂O released in 2 hr.

§It is arbitrarily assumed that 20% is released when 10% of the core vaporizes and that 5% is released when 2% of the core vaporizes

*Nitrogen atmosphere in cell is assumed to have 2 v/o oxygen

The FFTF hot cell described in BNWL-470⁽⁴⁰⁾ (pp. 2.11-6) is rectangular with dimensions 48 x 114 x 80 ft high. Its volume is 437,760 ft³ or 1.24 x 10¹⁰ cc. The floor area, A_F, is 5472 ft² or 5.08 x 10⁶ cm². In the present evaluation, we used the four vertical walls of the hot cell to represent A_w, the wall plating area, which is 25,920 ft² or 2.408 x 10⁷ cm². The ceiling area contribution was considered insignificant in these calculations.

In the case of the design basis accident, it is assumed that the excursion occurs very rapidly so that in effect there is no source time. In other words the mass appears in the hot cell instantaneously. Thus, Equation 7 can be rewritten

$$\frac{d\rho}{dt} = -\alpha\rho - \beta\rho - \gamma\rho \quad \dots(12)$$

These terms have the same definitions as before. The quantities α and β have the dimensions of the hot cell included in their numerical values. The combined relationships which were found to best fit the LTC data were $\Delta=0.5\mu$, and $r=0.2 + 0.7\rho^{0.5}$; "The prompt introduction of all the mass produces a maximum value of r at $t=0$. Thus the 0.2 does not influence matters at the start." However, $r_0=0.2$ also represents the final value of r , the mass median size of the smallest particle which remains airborne after the growth and fallout of the larger particles have occurred. If this value of r_0 were not included, the r value would approach zero as ρ does.

When materials with densities, d (gm/cc), other than that of sodium oxide are used, the velocity must be corrected to $v_s=0.012 dr^2$ cm/sec. If only sodium oxide is present, $v_s=0.027 r^2$. When, mixtures of Na₂O, $d=2.27$ and fuel consisting of PuO₂, $d=11.5$; UO₂, $d=10.9$; and possibly U₃O₈, $d=7.31$; then, the effective density must be estimated. As mentioned in Section VI the experimental determination of the relationships between r and ρ are all based on aerodynamic characteristics. These are (1) the deposition rate $dM_F/dt = \rho A_F v_s$, (2) the aerodynamic mass median radius $\sqrt{d} \cdot r$ and (3) the value of r_{50} , which is associated with T_{50} , the time for one-half of the mass to deposit on the floor. In the latter case, it is easy to show that $T_{50} = 0.69H/v_s = 0.69H/0.012dr^2$. Therefore, a reasonable estimate of the

effective particle density can be made by assuming complete homogeneity of the released mass such that the new agglomerated particle has an effective density

$$d_{\text{eff}} = \frac{M_{\text{UO}_2} + M_{\text{PuO}_2} + M_{\text{U}_3\text{O}_8} + M_{\text{Na}_2\text{O}}}{V_{\text{UO}_2} + V_{\text{PuO}_2} + V_{\text{U}_3\text{O}_8} + V_{\text{Na}_2\text{O}}}$$

where each V subscripted with a different material is the mass divided by its respective density. For convenience, all materials other than Na_2O have been assigned a density of 8 gm/cc; Na_2O has its nominal value of 2.27 gm/cc.

It is emphasized that the removal rate for floor settling is based on the product of dr^2 ; therefore, if a lower density is assumed, the settling velocity is reduced, hence the source term for site dose evaluation is conservative.

VIII. DISCUSSION OF RESULTS FOR FFTF HOT CELL

In Table 1, various estimates of the released mass of fuel, fission products, and sodium are shown. A convenient relation between the mass concentration $\rho_R = M_R/V$, in the FFTF hot cell and the released quantities of any material is $3.66 \mu\text{gm/cc}$ per 100 lbs of released mass, or $0.08 \mu\text{gm/cc}$ per Kg of released mass, since volume $V = 1.24 \times 10^{10} \text{cc}$. The volume of the hot cell is such that 1135 lb of Na can be oxidized per percent oxygen by volume in the hot cell. Therefore, the oxygen concentration of 2 v/o in the hot cell atmosphere specified by PNL can produce up to $112 \mu\text{gm/cc}$ of Na_2O ; for this reason cases A and C in Table 1 have the same density.

In programming the model for this study, the Adams-Moulton integration method was used to solve the equations. Equation (10) was programmed on the IBM-350/60 digital computer to provide the following output information.

- 1) Solutions of ρ/ρ_R vs time with $\gamma = 0, 0.1, 0.5$, and 1.0 day^{-1} , where ρ is the mass concentration which is airborne at any time, ρ_R is the initial instantaneous airborne mass concentration, and γ is the fractional leakage rate from the hot cell to the outer containment. The values of r , the effective monosize mass radius which remains airborne is also computed.
- 2) Solutions of the integral $V\gamma \int \rho dt / V\rho_R = M_L / V\rho_R$, where $M_L / V\rho_R$ is the mass fraction which leaks from the hot cell f_L . This ratio varies with time and with the values of γ . $V\rho_R = M_R$, the total mass of fuel, fission products, and sodium released into the hot cell.
- 3) Solutions of the integral

$$\frac{V \int \alpha \rho dt}{V\rho_R} = \frac{M_F}{V\rho_R}$$

which is the fraction of the material introduced into the hot cell which settles on the floor.

4) Solutions of

$$\frac{V \int \beta \rho dt}{V \rho_R} = \frac{M_w}{V \rho_R},$$

the fraction of the released mass which plates on the wall.

The runs were selected in order to show (a) the effect of the released mass concentration, ρ_R , on the fraction of the mass which has leaked, $f_L = M_L/M_R$, (b) the effect of the choice of the effective particle density d_{eff} on the fraction of mass leaked and (c) the effect of the leak rate on the fraction of mass leaked when both ρ_R and d_{eff} were varied.

The fraction, f_L see Table 2, of the original mass of material in the hot cell atmosphere which has leaked out can be used to estimate directly the fraction of PuO_2 or fission product mass which has leaked. The mass fractions of PuO_2 or fission products of the released material are represented by the constants f_P and f_{FP} respectively. Thus, $M_R \cdot f_L \cdot f_P =$ total mass of PuO_2 , and $M_R \cdot f_L \cdot f_{FP} =$ total mass of fission products which have leaked to the outer containment. Actually, $f_P M_R$ and $f_{FP} M_R$ are the total masses of these materials which are released. The quantity f_L increases in time to a maximum value for each leakage fraction, γ . The ratio of the mass of PuO_2 and FP's to the total material released is shown in Table 1 as f_P and f_{FP} respectively.

The activity in curies of a specific fission product isotope which has leaked is $f_L \cdot f_{FP} \cdot f_i \cdot f_n \cdot S$,

where: f_L = the fraction of all material which leaks and varies with time,

f_{FP} = the fraction, a constant, of the mass due to the fp's which are vaporized,

f_i = the fraction of the mass of fp due to species i ; f_i varies with time as the isotopes of that species decays,

f_n = the fraction of the mass of species i due to an isotope of mass number n ; f_n varies with time as the decay of an individual isotope occurs, and

S = the specific activity of the isotope, n .

TABLE 2
SUMMARY RESULTS OF ACCIDENT CALCULATIONS

Case	Run Number	Leakage Rate (γ day ⁻¹)	f_L at 2 hr (M_L/M_R)	$f_L^{(\infty)}$ (M_L/M_R)	ρ_R (μ gm/cc)	Particle Density d (gm/cc)	Hot Cell Envi- ronment	M_{LP} , Mass Pu Leaked (gm)	
								at 2 hrs	at $T=\infty$
A	4	0.1	.0037	.010				266	720
	6	0.5	.018	.056	75	2.3	N_2^*	1300	4030
	8	1.0	.036	.083				2600	5960
B	42	0.1	.00308	.0062			N_2^*	223	446
	43	0.5	.0157	.0332	27	8		1080	2390
	44	1.0	.031	.062				2230	4460
C	4	0.1	.0037	.010				266	720
	6	0.5	.018	.056	75	2.3	Air	1300	4030
	8	1.0	.036	.083				2600	5960
D	3	0.1	.0057	.0237			N_2^*	82	341
	5	0.5	.028	.125	22	2.3		403	1800
	7	1.0	.055	.18				790	2590
E	46	0.1	.0026	.0059				187	425
	47	0.5	.015	.0245	125	2.7	Air	1080	1760
	48	1.0	.0255	.0505				1836	3640
F	34	0.1	.00227	.00487	175	2.5	Air	164	351
	35	0.5	.0115	.0225				825	1620
	36	1.0	.022	.0417				1585	3000
G	37	0.5	.0085	.023				613	1655
	38	1.0	.018	.0434	265	2.3	Air	1300	3130

* N_2 atmosphere containing 2% oxygen

Since $f_{FP} \cdot f_i \cdot f_n \cdot M_R = M_n$, the mass of the isotope n produced at zero time; $f_L \cdot M_n = M_{Ln}$, the mass of isotope n which has leaked.

As previously discussed in Section 5, the present model assumes that all of the released materials are associated and that f_L can be interpreted as representing the ratio of the leaked quantity to the quantity which is released in the hot cell; thus,

$$f_L = \frac{M_L}{M_R} = \frac{M_{L FP}}{M_{FP}} = \frac{M_{LP}}{M_P} = \frac{M_{Li}}{M_i} = \frac{M_{Ln}}{M_n} = \frac{N_{Lc}}{N_c},$$

where M_R is the total mass of all materials; M_{FP} , the total mass of fission products; M_P , the total mass of PuO_2 ; M_i , the total mass of the fission product species ($z = \text{constant}$); M_n , the total mass or number of moles of the isotope ${}_z^n(FP)$; and N_c , the total number of curies of a group of fission products ($z = \text{constant}$) or of an individual fission product ${}_z^n(FP)$.

The number of curies of an individual isotope, a species, or a group of isotopes, such as volatiles solids, or halogens all vary as a function of time; therefore, the value of f_L at a given time should be multiplied by the number of curies present at the time of interest.

Another method for obtaining the quantity of a material or the number of curies that leak from the hot cell is to determine an effective removal term from the plots of ρ/ρ_R vs time at a given leak rate. The effective removal constant is then programmed into radiological site dose codes such as COMRADEX⁽⁴¹⁾. As the slope of ρ/ρ_R changes with time, a different removal constant should be obtained. Thus, a series of values which vary in time can be used in codes previously programmed for constant removal rates.

Table 2 presents the code case numbers for the accidents described in Table 1. Figures 4-8 present the time behavior of M_L/M_R and ρ/ρ_R for $\gamma = 0.1$ per day. Accidents A and C were computed with a conservative estimate of material density, d , through a programming error; however, the effect on the results was negligible. A composite curve showing M_F/M_R , M_w/M_R , M_L/M_R , and ρ/ρ_R vs time is also shown in these figures. Two curves (Cases A to E) [$\gamma = 0.1 \text{ day}^{-1}$] of ρ/ρ_R are presented, one shows the early dependence with time and another shows ρ/ρ_R for longer periods of time. The curves

Figure 4a. Case A or C, $\gamma = 0.1/\text{day}$, $d = 2.3 \text{ gm/cc}$, $\rho_R = 75 \text{ } \mu\text{gm/cc}$

Figure 4b. Case A or C, $\gamma = 0.1/\text{day}$, $d = 2.3 \text{ gm/cc}$, $\rho_R = 75 \text{ } \mu\text{gm/cc}$

Figure 4c. Case A or C, $\gamma = 0$ 1/day, $d = 2.3$ gm/cc, $\rho_R = 75$ μ gm/cc

Figure 4d Case A or C, $\gamma = 0.1/\text{day}$, $d = 2.3 \text{ gm/cc}$, $\rho_R = 75 \mu\text{gm/cc}$

Figure 5a Case B, $\gamma = 0$ 1/day, $\rho_R = 27$ $\mu\text{gm/cc}$, $d = 8$ gm/cc

Figure 5b Case B, $\gamma = 0$ l/day, $\rho_R = 27$ $\mu\text{gm/cc}$, $d = 8$ gm/cc

Figure 5c. Case B, $\gamma = 0.1/\text{day}$, $\rho_R = 27 \mu\text{gm/cc}$, $d = 8 \text{ gm/cc}$

Figure 5d. Case B, $\gamma = 0$ 1/day, $\rho_R = 27$ $\mu\text{gm/cc}$, $d = 8$ gm/cc

Figure 6a Case D, $\gamma = 0$ 1/day, $\rho_R = 22$ $\mu\text{gm/cc}$, $d = 2.3$ gm/cc

Figure 6b. Case D, $\gamma = 0.1/\text{day}$, $\rho_R = 22 \mu\text{gm/cc}$, $d = 2.3 \text{ gm/cc}$

Figure 6c. Case D, $\gamma = 0.1/\text{day}$, $\rho_R = 22 \mu\text{gm/cc}$, $d = 2.3 \text{ gm/cc}$

Figure 6d Case D, $\gamma = 0$ 1/day, $\rho_R = 22$ $\mu\text{gm/cc}$, $d = 2.3$ gm/cc

Figure 7a. Case E, $\gamma = 0$ 1/day, $\rho_R = 125$ $\mu\text{gm/cc}$, $d = 2.7$ gm/cc

Figure 7b Case E, $\gamma = 0.1/\text{day}$, $\rho_R = 125 \mu\text{gm/cc}$, $d = 2.7 \text{ gm/cc}$

Figure 7c. Case E, $\gamma = 0.1/\text{day}$, $\rho_R = 125 \mu\text{gm/cc}$, $d = 2.7 \text{ gm/cc}$

Figure 7d. Case E, $\gamma = 0.1/\text{day}$, $\rho_R = 125 \mu\text{gm/cc}$, $d = 2.7 \text{ gm/cc}$

FFTF 34

7908 85
0200 0210

Figure 8a. Case F, $\gamma = 0.1/\text{day}$, $\rho_R = 175 \mu\text{gm/cc}$, $d = 2.5 \text{ gm/cc}$

Figure 8b. Case F, $\gamma = 0.1/\text{day}$, $\rho_R = 175 \mu\text{gm/cc}$, $d = 2.5 \text{ gm/cc}$

Figure 8c. Case F, $\gamma = 0$ 1/day, $\rho_R = 175$ $\mu\text{gm/cc}$, $d = 2.5$ gm/cc

Figure 8d. Case F, $\gamma = 0$ 1/day, $\rho_R = 175$ $\mu\text{gm/cc}$, $d = 2.5$ gm/cc

representing the time and space distribution of the mass when $\gamma = 0.5$ and 1.0 are available, but have not been included in the present report.

Table 2 also presents the mass of plutonium oxide present in the high bay at the end of 2 hr and at $t = \infty$. The limiting value for $t = \infty$, is approached in a very short time (always less than 1 day). This time can be compared to that which would occur if there were no removal terms; i. e., if leakage alone occurred.

The time for $f_L = 0.97 f_L^{(\infty)}$, with no fall-out, (i. e. after 5 half times of out-leakage) may be computed as $5 \times 0.693/\gamma = 35, 7$, and 3.5 days for $\gamma = 0.1, 0.5$, and 1.0 per day respectively.

One may also compare the leakage fraction f_L at 2 hr with and without fall-out. Values of $f_L = M_L/M_R$ (2 hr) with fall-out are listed in Column 3 in Table 2. The corresponding value without fall-out may be approximated by: $\gamma \text{ 1/day} \times 1/12 \text{ day}$. Thus, for $\gamma = 0.1, 0.5$, and 1.0 per day $f_L = 0.008, 0.04$, and 0.08 respectively. By comparing the tabulated f_L values with the approximated numbers above, it becomes clear that the benefit from agglomeration and fall-out is not great at 2 hr (a factor of 2 to 4); however, at $t = \infty$, one must compare the values in Column 4 with a value of $f_L = 1.0$. Thus, the benefit now becomes a factor of 200 to 5, dependent on the leak rate and case number considered.

Case G in Table 1 and 2 represents the conditions following the DBA where all of the sodium of the core is assumed to be expelled from the reactor vessel. The released sodium is assumed to rapidly form a pool fire configuration* over the entire hot cell floor area and to burn at a rate of 10 lb/hr-ft^2 in an air environment. Approximately $1/5$ of the oxidized sodium is released, and 12 tons of sodium is oxidized in 2 hr.

All of the oxygen in the hot cell is consumed at a rate equivalent to a half time of 19 minutes, or with a time constant of $0.69/19$ minutes. Figures 9 and 10 show the computed parameters previously mentioned, all as a

*If all of the sodium in the pool above the core were expelled, one would expect that a major fraction of the liquid would be in a state of subdivision which would be far removed from a "spray" or "drop" configuration, and would, for the most part, fall back to the hot cell floor rapidly to form a pool.

FFTF 37

7908-86
0004 0004

Figure 9a. Case G, $\gamma = 0.5/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm/cc}$

Figure 9b. Case G, $\gamma = 0.5/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm/cc}$

Figure 9c. Case G, $\gamma = 0.5/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm/cc}$

Figure 9d Case G, $\gamma = 0.5/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm/cc}$

FFTF 38

7906-66
0011 0011

Figure 10a. Case G, $\gamma = 1.0/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm, cc}$

Figure 10b. Case G, $\gamma = 1.0/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm/cc}$

Figure 10c. Case G, $\gamma = 1.0/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm/cc}$

Figure 10d. Case G, $\gamma = 1.0/\text{day}$, Source Time = 2 hr, $\rho_R = 240 \mu\text{gm/cc}$,
 $d = 2.3 \text{ gm/cc}$

function of time, when $\gamma = 0.5$ and 1.0 per day. It was assumed that the pressures generated by the fire would produce a higher leak rate; therefore, no computations were made for $\gamma = 0.1$ per day. The source time, τ , was chosen as 2 hr and a linear approximation was made for the source term S_M (see Equation 7).

By comparing Cases F and G where $\rho_r = 175$ and $240 \mu\text{gm/cc}$ respectively it becomes apparent that introduction of a source time of 2 hr did not appreciably affect the mass of PuO_2 which leaked out.

The above discussion concerning the total mass and the mass of a specific material that leaks, also applies to the mass fraction that plates or settles.

IX. CONCLUSION AND RECOMMENDATION

The present evaluation is based on an empirical mass model which relates the particle size distribution of a given mass of material to the mass concentration which is present. The model assumes that all materials other than gases will agglomerate and grow to sizes which can be represented by an equivalent mass radius. All of the materials which are airborne agglomerate to particles which are characterized as having a total mass composed of the mixture which is airborne. The effective size of the aggregate is proportional to the total mass of material which is airborne.

In these accident studies, a constant mass of fuel is vaporized (10% of the core for a 3000-Mw-sec energy release) but the amount of sodium which is released varies from 0 to 40,000 lbs; therefore, the total mass of material which is airborne increases. It is apparent from Table 2 that the leaked fraction f_L , decreases as the mass concentration increases. The relation of f_L at $t=\infty$ to ρ_R may be stated algebraically, based on code results as

$$f_L(\infty) = \frac{5.0 \gamma}{(\rho_R d)^{0.8}} \quad \dots (13)$$

One can see that increasing the mass concentration will indeed lower the leak fraction.

The above Equation (13) is the most significant result of this study. Since the mass of PuO_2 is constant, the leaked PuO also decreases as ρ_R increases. It can be shown that the mass concentration of any material in the inner containment vessel, ρ_a , and the leaked mass, M_{La} , are functions of the total initial concentration in the hot cell, ρ_R .

$$\frac{M_{La}}{V_2} = \frac{5.0 \gamma f_a}{(d)^{0.8}} \frac{\rho_R^{0.2} V_1}{V_2} = \frac{5.0 \gamma \rho_a V_1}{(d)^{0.8} (\rho_R)^{0.8} V_2} \quad \dots (14)$$

where f_a is the fraction of any material (released in) the hot cell, and V_1 and V_2 are the volumes of the hot cell and outer containment vessel.

If the knowledge of ρ_R (total mass concentration released) is uncertain, there will be a corresponding uncertainty in the knowledge of the mass "a" released, even though ρ_a is well known. The more mass (sodium, fuel, structure) one is able to assume released, the more conservative will the estimate become of the mass of Pu released (or any other toxic entity).

If one attempts to determine the radiological dose received because of leakage from the outer containment, the fraction f_L must be used to compute the inventory of the materials which will leak. Consider Case E, of Table 2, for a leakage of 0.1 per day at 2 hr after the accident. There would be the following inventory of materials available for leakage from the secondary containment vessel.

PuO ₂ , 187 gm	= 3. x 10 ³ C _i
Halogens	4.15 x 10 ⁴ C _i
Vol Solids	2.32 x 10 ⁴ C _i
Noble Gas	5.42 x 10 ⁵ C _i

For the noble gases, 100% are assumed to be released, and their leak fraction is $\gamma \times 2/24$. Of the total inventory of the halogen and volatile solids, 20% was released; multiplied by $f_L = 0.0026$ (see Table 2). For the core, 10% of the PuO₂ was assumed to be vaporized and released, and multiplied by $f_L = 0.0026$.

The reduction factor attributable to agglomeration and fallout for the 2-hr leak fraction is $0.00835/0.0026 = 3.22 = \gamma \times 2/24/f_L$ (2 hr) and the factor is the same for PuO₂, halogens, and volatile solids. If a higher fraction of PuO₂ or fission products is assumed to vaporize (than those suggested in Table 1) multiply the computed values of f_L by the fraction assumed to be released from the fuel. No significant error in the mass leaked will occur.

The relationship derived from this study for f_L at 2 hr and the released mass is f_L

$$f_L \quad (2 \text{ hr}) = 0.17 \left(\frac{5}{d} \right)^{0.46} \frac{\gamma}{(\rho_R)^{0.46}} \quad \dots (1b)$$

where d is the material density and ρ_R , as before, is the total released mass concentration in the cell. This relationship is somewhat inaccurate, but is conservative at $\rho_R = 100 \text{ } \mu\text{gm/cc}$ and for $d = 5 \text{ gm/cc}$. The exponents are 0.8 for d and ρ_R in the expression for f_L at $t=\infty$. A calculation of f_L for times short compared to a characteristic time for the hot cell (the characteristic time is of the order of 1 to 2 hr) would yield the result $f_L = \gamma t$; i.e., f_L would in this case show no dependence on ρ_R and d . For times like 2 hr, comparable to the characteristic time, f_L shows the intermediate form of dependence with the exponents of d and ρ_R equal to 0.46.

The time for 50% of the total mass that will leak T_L (Sec) which was determined for the FFTF study ($\gamma = 0.1$ per day) was

$$T_L = \frac{2.16 \times 10^4 \text{ (Sec)}}{(\rho_R)^{0.46} \times (d)^{0.79}} \quad \dots (16)$$

where ρ_R and d are as previously defined.

The time to reach approximately 97% of the equilibrium leak fraction is approximately 5 days for all of the cases studied when $\gamma = 0.1$ per day. As γ increases to 1.0 per day, the time to leak 97% of the leaked mass decreases to ~ 3 days, and the value of T_L is less than that in Equation (16).

For a complete study of the radiological dose delivered at a particular distance downwind from the reactor complex, one still needs to use the usual techniques employed in a Site Dose Code. For this reason, values of ρ/ρ_R , the ratio of the released mass which is airborne to the total released material is included in the report. These curves allow one to choose linear approximations for a given leak fraction which can be programmed in the Site Dose Code as a removal factor for each fission product or fuel material.

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