THE EFFECT OF AEROSOL AGGLOMERATION ON THE
REDUCTION OF THE RADIOLOGICAL SOURCE TERM
FOR THE LMFBR DESIGN BASIS ACCIDENT

BY

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ABSTRACT

Calculations of the stirred settling rate of particles with heterogeneous distributions characterized as log normal are presented. The data are presented in a form which allows one to estimate the fractional decrease of the initial concentration versus time for a given distribution. A log-normal distribution is completely characterized by the median size and geometric standard deviation. Additional data are presented which allows one to determine the leaked fraction of the mass which is released into a vessel as a function of time for a given distribution. Finally, an empirical relation is suggested for estimating the median particle size produced by agglomeration of initial particles as a function of the mass introduced into the test chamber. The settling velocities are computed on the base of Stokes Law.

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Figure 1. Containment Features of 500-Mwe Plant

I. INTRODUCTION

Studies are currently being conducted at Atomics International^{*,} to characterize the release and transport of effluents generated during major Liquid Metal Fast Breeder Reactor (LMFBR) accidents. The information developed under this program and others,⁽¹⁻³⁾ see Section III, supports a realistic assessment of the radiological source term for the most severe fast reactor accidents.

The Design Basis Accident (DBA) postulated for a 500-Mwe LMFBR⁽⁴⁾ produces an estimated gross energy release of $\sim 13,000$ Mw-sec of which some 2600 Mw-sec is available to do work. This explosive force disperses some of the vaporized (3500°C) fuel mixture into the upper sodium pool and then sprays the mixture up into the cover gas, see Figure 1. The vaporized sodium-fuel mixture vents into V_1 , the primary vaults, where it is available to leak ($\sim 10\%$ /day at 10 psi) into V_2 , the secondary containment space. Some of the airborne sodium is oxidized to form spherical Na_2O aerosol particulates, and the vaporized UO_2 - PuO_2 is oxidized in air to form supercooled spheres which link up to form branched-chain aggregates. Both the fuel and sodium oxide aerosols[†] spontaneously and continuously coagulate into larger particles (cf Figure 2a). When both aerosols are formed simultaneously, the sodium oxide agglomerates onto the fuel aerosol as well, forming a composite branched-chain aerosol, cf Figure 2b. In the agglomeration process, the aerosols which come into contact stick together with almost 100% efficiency. This decreases the number of particles and increases the effective particle radius. As the aerodynamic radius of the aerosol becomes larger the particle settles faster. For high concentrations, large particles are produced, and the settling rate is much faster than the rate of removal by leakage for typical leak rates; thus, after a very short time, the material available for leakage is considerably reduced.

The present study is concerned with the development of a computational technique which aids in the determination of the source term for the LMFBR DBA radiological dose at the reactor site. The ultimate source term which

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†The aerosols from arc-vaporized UO_2 -SS mixtures and burning sodium have been studied at AI; ORNL has investigated arc-vaporized preirradiated UO_2 and other studies at BNL show that PuO_2 aerosols behave similarly to those of UO_2 .

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Figure 2a. Branched Chain Coagulation of U_3O_8 Aerosols

directly controls the site selection and boundaries is the quantity of airborne radioactive material which leaks from V_2 , the outer containment. The radioactive mass which escapes from the outer containment is assumed to be a function of the gas leak rate from the primary vaults, and the agglomeration, plating, and settling characteristics of the aerosol.

The computational procedure for determining the specific reduction factor for the source term is based on experimental and analytical studies conducted at AI which are described and referenced within the body of the report. In these studies, all growth and settling of the accident-produced aerosol is assumed to occur in the primary containment system, and no credit is taken for settling of particles after they have leaked from the primary vaults, or for depletion of the particles by impaction and settling during their flow through the leak paths.

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Figure 2b. Coagulation of U_3O_8 and Na_2O Aerosols
Generated Simultaneously

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II. REVIEW OF AEROSOL PHYSICS

A. AEROSOL COAGULATION LAW

Whytlaw-Gray (1932) and his co-workers at Leeds⁽⁵⁾ have shown that the coagulation of many aerosols 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

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

where K is the coagulation constant. Thus, under conditions of little fallout and plateout, 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.

By differentiating Equation 1 one can see that the rate at which particles disappear by agglomeration depends on the square of the number of particles present and the constant K .

Smoluchowski's equation⁽⁶⁾ for a monosize aerosol:

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

Thus, coagulation is very rapid in the early stages when the number concentration is high, and later it falls off rapidly. A comparison of the value of the coagulation constant obtained experimentally for many different aerosols in air under normal conditions of temperature and pressure indicates that although K varies with a particular smoke, coagulation does not depend very markedly on the nature of the aerosol.

The rapidity of coagulation as a function of concentration is better appreciated when the time, T_{50} , to halve the original number of particles is calculated.

*Equation 1 is the solution of Smoluchowski's^(6,7) equation for a monosize aerosol (Equation 3). It is also of the same form as the solution of the general coagulation equation of a heterogeneous aerosol for agglomeration only.⁽⁸⁾

This is found from Equation 1 by letting $n = n_o/2$, then the agglomeration half-time is, $T_{50} = 1/n_o K$; i.e., the half-time is proportional to $1/n_o$. Taking the value of K for an aerosol as $5.0 \times 10^{-10} \text{ cm}^3/\text{sec}$, we find that T_{50} for a system of 10^{11} particles per cm^3 , is: 0.02 sec; for 10^6 per cm^3 , 33 min; and for 10^5 per cm^3 , 5.5 hr. Therefore, when the number concentration is high, coagulation is so rapid that the number of particles remaining after a few minutes is practically independent of the initial number. The average size of the particles which remain also changes as agglomeration occurs.

If the particles are assumed to be monosize spheres, then $C_R = (4/3)\pi r^3 \rho n$ where ρ is the particle density and C_R is the mass concentration. The particle radius, r is related by Equation 1 and C_R as:

$$\frac{4\pi\rho(r^3 - r_o^3)}{3C_R} = Kt \quad \dots(3)$$

For a fixed time when $r_o \ll r$, and for a monosize distribution:

$$r \sim (C_R)^{1/3} \quad .$$

B. STIRRED SETTLING LAW FOR MONOSIZE AEROSOLS⁽⁹⁾

The mass concentration, dC , of a monosize aerosol ($\sigma = 1$) that settles during a time, dt , is proportional to the concentration C at that time. The fraction of particles with velocity of fall v , that settle out of a room of height, H , in time, is

$$\frac{dC}{dt} = -\frac{v}{H} C \quad \dots(4)$$

By integrating this equation, we find that

$$C = C_o \exp(-vt/H) \quad , \quad \dots(5)$$

where C_o is the initial concentration (equivalent to $C_{\max}$). Setting $C = 1/2(C_o)$ one can define

$$\frac{T_{50}}{H} = \frac{0.69}{v} \quad , \quad \dots(6)$$

where :

$v_{(\text{cm/sec})} = 0.013\rho'(r_{50})^2$ = Stokes law for a particle of size, r , settling in air,

r_{50} = equivalent monosize Stokes radius in microns and the mass median radius, and

ρ' = effective density of the agglomerated aerosol.

We can apply these monosize ($\sigma = 1$) stirred settling relations to the AI experiments ($\sigma = 1.5$ to 2.5 , see Section III) by treating the mean of the distribution as if it obeys the monosize relation at the half-time. By adding a term to Equation 2 to account for particle removal by fallout, the problem reduces to the solution of the following simultaneous equations for monosize particles when both Brownian motion agglomeration and settling occur.

From Equation 2,

$$\frac{dn}{dt} = -Kn^2 - \lambda n + S_n \quad \text{and} \quad \dots(7)$$

$$\frac{dC}{dt} = -\lambda C + S_v, \quad \dots(8)$$

where:

K = agglomeration constant,

S = source term, S_n is the number of particles/cm³-sec and S_v is the mass concentration/sec,

λ = removal constant = $v(r)/H$,

H = height of container in cm,

n = number of particles/cm³, and

ρ = effective density of the particles.

In order to solve these equations, one can make use of the proportionality of C and n as before;

$$\begin{aligned} C &= (4/3)\pi r^3 n \rho, \\ r^3 &= (3/4)C(1/\pi)(1/n)1/\rho, \quad \text{or} \\ r &\approx (C/n)^{1/3}. \end{aligned}$$

In a fixed geometry for a specific C and r , the growth of the particles would be limited, since after they reached a certain size, their settling rate would become so large that they would be removed from the chamber as contrasted to Equation 1, the simple Whytlaw-Gray relation, where the particles grow forever. It should also be noted that the time to reach a given size (or the size which is reached in a given time) varies with the mass concentration. If both settling and agglomeration occur in a given size chamber, a maximum particle size should be produced for a given initial concentration and height.

The simple expressions described above represent only one mechanism of agglomeration. For highly dispersed particle distributions, gravitational settling of different sizes accounts for even higher agglomeration rates.

III. SUMMARY OF AEROSOL EXPERIMENTS*

A. LTC EXPERIMENTS

The characteristics of both sodium and UO_2 aerosols have been investigated at AI in a laboratory test chamber, LTC, (40 ft³ volume, 6 ft high), by burning a pool of liquid sodium in various oxygen environments (producing Na_2O aerosols), or by arc-vaporization of UO_2 in stainless steel containers in an air environment (producing Fe_3O_4 and U_3O_8 aerosols). In this series of experiments, the released mass concentration, C_R , was varied from 2 to 60 $\mu\text{gm}/\text{cm}^3$ of Na_2O and from 0.1 to 20 $\mu\text{gm}/\text{cc}$ of U_3O_8 & Fe_3O_4 . Following the release of material into the chamber, sequential samplers⁽¹⁷⁾ automatically collect materials which plate on the walls, settle on the floor, or remain in the air. Samples of the particle distribution in the air are obtained both with round jet cascade impactors^(14,15,17) and electrostatic precipitators. Cumulative samples are collected on the walls and floor, and a mass balance is obtained throughout the experiment; the sum of the material on the wall and floor and that remaining airborne is equal to the total mass released. Deposition samplers used at any distance above the floor have been shown to collect equal amounts of mass in a given time period after the aerosol formation, and thus demonstrate uniform "stirred" concentration conditions in the chamber. In these tests, the floor and wall deposition rates and the airborne concentration allow one to make an estimate of the effective particle size being deposited as a function of time.

There are several phenomena which conveniently characterize all of the LTC aerosol experiments. The following parameters (See Figure 3 for a schematic diagram to aid in the following discussion.) show a dependence on the released mass concentration which is independent of the chemical specie as follows:

- 1) The ratio $M_F(\infty)/M_R$ increases as C_R increases, and implies that larger particles are produced at higher concentrations. M_R is the released mass from the burning sodium.
- 2 The wall deposition rate increases with increasing airborne concentration because higher concentrations are associated with higher temperature gradients to the walls (thermophoretic deposition). The wall plating half-time, T_{50} , decreases as C_R increases.

*Please see References 1 through 3 and 10 through 17.

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Figure 3. Diagram of Nomenclature Used in Experimental Descriptions

where

$$M_R = M_F(\infty) + M_w(\infty)$$

$$\frac{M_R}{V} = \bar{C}_R, \quad C_R \geq C_o \text{ as a function of } C_R \text{ and } \tau.$$

C_o = Maximum airborne mass concentration

T_{50} = Time for C_o to decrease by one-half

T_{50F} = Time for 1/2 of the mass to deposit on the floor after τ

τ = Time to introduce C_R into the chamber

- 3) The ratio C_o/C_R decreases as C_R increases and suggests that there is growth of the initial distribution during the production time, τ .
- 4) The initial size distribution of the released spherical sodium oxide aerosol is log normal, with an initial mass median radius, $r_{50(o)}$, of 0.54μ and a geometric standard deviation, σ , of 2.1. The initial mass median radius of the U_3O_8 - Fe_3O_4 branched chain is 0.3 to 0.5 with a geometric standard deviation of ~ 2 . $r_{50(o)}$ is not a function of C_R . If $C_R \ll 1\mu\text{g/cc}$, r_o may not increase but only decreases as settling and plating occur; i.e., little agglomeration occurs.
- 5) $T_{50}(F)$ is the time for one-half of the mass to be deposited on the floor after production has ceased. As C_R increases, $T_{50}(F)$ decreases. $T_{50}(F)$ decreases as the size of the particles increase. The median particle size increases to larger sizes as the mass of released material, M_R , increases regardless of the shape of the aerosol aggregate, branched-chains or spheres, U_3O_8 or Na_2O ; see Figure 3.
- 6) In all of the experiments, the initial median radius changes in time to a value, r_{max} . The time for r_{max} to occur decreases as C_R increases and, τ , the production time decreases. r_{max} is the maximum mass median particle radius to which the initial distribution grows. There are three methods for characterizing r_{max} at AI. These are: (1) r_m , from the floor deposition rate, (2) $r_{50}(F)$, from the floor deposition half-time, $T_{50}(F)$, and (3) r_a , from measurements of the airborne particle size distribution obtained by using cascade impactors. All three estimates of r_{max} confirm that growth is occurring; see Figure 4.
- 7) Mixtures of aerosols produced simultaneously by arc vaporizing UO_2 -stainless steel in air and by burning liquid sodium were also studied. Both materials agglomerated together to form a complex branched aggregate which settled to the floor at a rate proportional to the combined mass concentration. Figure 5 shows a comparison of the initial floor deposition half-times vs the released concentration of sodium oxide and various mixtures of U_3O_8 , Fe_3O_4 , and Na_2O .

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Figure 5. Released Material vs Floor Deposition Half-Time

- 8) All measurements of the particle size distributions are fitted to a log normal distribution. The value of σ , the geometric standard deviation, varies in either UO_2 or Na_2O experiments from initial values of 2 to 2.5 to final values of 1.5 to 1.7.

B. EMPIRICAL MODEL DEVELOPMENT

Aerosol studies performed on irradiated UO_2 at ORNL and on UO_2 , PuO_2 , and Na_2O at BNL also show that as the initial airborne concentration increases there is a decrease in the time for the initially observed concentration to be decreased by one-half.⁽¹⁾

The curves shown in Figure 5 indicate that the initial half-time, $T_{50}(F)$, of the floor deposit is inversely related to the released mass concentration, C_R . The empirical fit to the Atomics International floor settling data* in Figure 5 are:

For U_3O_8 & Fe_3O_4 aerosols,

$$\frac{T_{50F}}{H} = \frac{40/3}{(C_R)^{0.6}} \text{ (sec)} \quad , \quad \dots(9)$$

and for Na_2O aerosols,

$$\frac{T_{50F}}{H} = \frac{70/3}{(C_R)^{0.8}} \text{ (sec)} \quad , \quad \dots(10)$$

where:

H = chamber height (cm) and
 $C_R > 1\mu\text{gm/cm}^3$.

These empirical equations can be developed into a model based on the stirred settling of monosize aerosols.⁽⁹⁾ From Equations 6 and 9 one can write for U_3O_8 aerosols:

$$\frac{0.69}{0.012\rho r_{50}^2} = \frac{40/3}{C_R^{0.6}} \text{ or } \sqrt{\rho} r_{50} = K_1 C_R^{0.3} = R_{50} \quad . \quad \dots(11)$$

*The floor settling data is used in preference to the air concentration data since the former is influenced only by Stokes law.

For Na_2O aerosols, Equations 6 and 10 can be combined to give:

$$\frac{0.69}{0.012 \rho r_{50}^2} = \frac{70/3}{C_R^{0.8}} \quad \text{or} \quad \sqrt{\rho} r_{50} = K_2 C_R^{0.4} = R_{50} \quad \dots (12)$$

The values of K_1 and K_2 are 2.1 and 1.6, respectively.

Both sets of data can be fitted reasonably well for a relation which is independent of chemical species:

$$\frac{T_{50F}}{H} = \frac{(50 - 70)/3}{C_R^{0.78}} \quad \text{and} \quad \sqrt{\rho} r_{50} = (1.9 \text{ to } 1.6) C_R^{0.39} = R_{50} \quad \dots (12a)$$

IV. DEVELOPMENT OF A MODEL FOR STIRRED SETTLING OF A HETEROGENEOUS DISTRIBUTION OF AEROSOLS

The distributions measured in the LTC experiments were fitted to log normal form. The initial geometric standard deviation, σ , varied from 2 to 2.5 down to values of 1.5 to 1.7 for σ as the airborne concentration decreased; however, variations in the value of σ had little effect either on the relationship of the initial $T_{50}(F)$ to C_R or on the empirical relation of r_{50} to C_R computed in Equations 11 and 12.

In order to test this observation that the above relations are largely independent of σ , it is necessary to compute the stirred settling half-times of heterogeneously distributed aerosols as a function of σ . To aid in this analysis, we begin with the stirred settling law for monosize aerosols. To Equation 4, we add a leakage term in order to make the analysis appropriate for reactor hazard studies: for $\sigma = 1$

$$\frac{dC}{dt} = -\left(\frac{v}{H}\right) \cdot C - \left(\frac{Q}{V}\right) \cdot C \quad \dots(13)$$

The solution of this equation is:

$$C = C_o \exp \left[-\left(\left(v + \frac{Q}{V} H \right) \frac{t}{H} \right) \right] \quad \text{for } \sigma = 1, \quad \dots(14)$$

where:

C_o = initial concentration and is the equivalent of C_R if τ , the production time, is very short compared to the initial half-time, and

$Q/V = \gamma(\text{sec}^{-1})$ = fractional leakage from the primary vaults.

If $\gamma = 0$, we define $C/C_o = f_R$, and define

$$f_A = \frac{C}{C_o} = \exp \left[-\left(v + \frac{Q}{V} H \right) \frac{t}{H} \right] \quad \text{when } \gamma > 0.$$

We wish to derive the equivalent of Equation 14 for a heterogeneous distribution.

The heterogeneous, $\sigma > 1$, logarithmic probability distribution function is⁽¹⁸⁾

$$f(r) dr = \frac{1}{\sqrt{2\pi} \ln \sigma} \exp \left[-\frac{\ln^2(r/r_g)}{2 \ln^2 \sigma} \right] d(\ln r) \quad , \quad \dots(15)$$

where:

$f(r)dr$ = number of particles between r and $r + dr$,

$r_g(o)$ = geometric mean radius = $r_{50(o)} e^{-(3 \ln^2 \sigma)}$,

r = particle radius, and

σ = geometric standard deviation.

The number of particles, $N(r,t)$ with radii between r and $r + dr$ at time, t , from Equation 14

$$N(r,t) dr = N_o f(r) \exp \left[-\left(v(r) - \frac{Q_H}{V_H} \right) \frac{t}{\tau} \right] dr \quad , \quad \dots(16)$$

where:

N_o = total number of particles of all radii at time, $t = 0$.

The total mass of particles is proportional to the total volume of the particles and can be related at $t = 0$ as follows:

$$M_o = \frac{4}{3} \pi \rho N_o \int_0^\infty r^3 f(r) dr = N_o \frac{4}{3} \pi \rho r_g^3 \exp \left(\frac{9}{2} \ln^2 \sigma \right) \quad . \quad \dots(17)$$

This integration, shown in Equation 17, yields the familiar volume average particle radius cubed:⁽¹⁹⁾

$$\overline{r_v^3} = r_g^3 \exp \left(\frac{9}{2} \ln^2 \sigma \right) \quad . \quad \dots(18)$$

The ratio of the mass of particles at time, t , to the mass of particles at time, 0, from Equation 17 is:

$$\frac{M(t)}{M(o)} = \frac{\int_0^\infty r^3 N(r,t) dr}{N_o r_g^3 \exp \left(\frac{9}{2} \ln^2 \sigma \right)} = \frac{C(t)}{C(o)} \quad . \quad \dots(19)$$

Substituting from Equation 16 into Equation 19, we see that the fractional amount remaining airborne, f_A is

$$= \frac{\exp\left[-\frac{Q}{V}H\left(\frac{t}{H}\right)\right] \left\{ \int_0^\infty d\ln\left(\frac{r}{rg}\right) e^{-v(r)t/H} \cdot \frac{r^3}{\sqrt{2\pi}\ln\sigma} \left(-\frac{\ln^2 r/rg}{2\ln^2\sigma}\right) \right\}}{rg^3 \exp\left(\frac{9}{2}\ln^2\sigma\right)} \quad \dots(20)$$

If the $\exp\left(-\frac{Q}{V}t\right)$ is omitted, Equation 20 becomes the fraction, f_R , remaining airborne in a completely closed system; i.e., no leakage.

The fractional mass, f_L , that leaks from the primary vaults into a second compartment is:

$$f_L = \frac{\gamma H \int_0^{T/H} e^{-(\gamma H)t/H} d(t/H) \int_0^\infty d\ln\left(\frac{r}{rg}\right) e^{-v(r)t/H} \cdot r^3 \exp\left(-\frac{\ln^2 r/rg}{2\ln^2\sigma}\right)}{\sqrt{2\pi} \cdot \ln\sigma \cdot rg^3 \exp\left(\frac{9}{2}\ln^2\sigma\right)} \quad \dots(21)$$

Equations 20 and 21 have been programmed for solution on the IBM S/360-50 Digital Computer. The code uses the log normal distribution, Equation 16, and computes the fraction of the released mass remaining in suspension and the fraction of the released mass which leaks. In the solution, $v(r)$ was solved as the Stokes velocity with the Cunningham correction.

$$v(r) = 1.2 \times 10^{-2} \rho \cdot r^2 \left[1 + \frac{1}{r} (0.083 + 0.0264 \exp(-16.7r)) \right] .$$

All solutions are obtained for a material density, ρ of 1 gm/cm^3 . Thus, $R_{50} = \sqrt{\rho} r_{50} = \text{AER}$, the aerodynamics radius and $R_{50}^2 = \rho r_{50}^2$. Equation 20 computes the fraction of the released material which remains in suspension. Solutions were made in terms of t/H (sec/cm) for $\gamma H = 0$. It was desired to study, r_{50} , the mass median radius as a function of t/H . The functional relation was obtained for a series of values of $C/C_o = f_R$, the fractional amount remaining airborne when there is no leakage. Since ρ was 1 gm/cm^3 , the abscissas are labeled R_{50} .

The solutions are shown in graphical form in Figures 6 through 13. Each figure represents a given value of f_R and shows how t/H changes with R_{50} . There are four curves for each figure and each curve represents a different σ .

In Figure 8, R_{50} vs t/H is shown when $C/C_o = 0.5 = f_R$. The definition of $T_{50}(F)$ for the experimental data in LTC is equivalent to the relation plotted in Figure 9. One should note that the magnitude of σ does not have a great deal of effect on the value of t/H for a given r_{50} in microns, when the fraction f_R is 0.5.⁽¹⁹⁾ For the family of curves in Figure 8, one can write a general relation as:

$$\frac{50}{r_{50}^2} = \frac{T_{50}}{H} \text{ (sec/cm)} = \frac{50}{R_{50}^2}, \quad \dots (22)$$

which is independent of the geometrical standard deviation, σ . If Equation 22 is equated to the observed relationship shown in Equations 9 and 10, one can see that the relations are not too different from those estimated in Equations 11 and 12 having the monosize relationships. This may be fortuitous, for different height chambers may have other relationships to C_R , when agglomeration occurs.

Solutions of Equation 21 were used to compute the leaked fraction, f_L , of the released mass as a function of r_{50} for various values of γH and t/H . The values of γH were equal to 1.0×10^{-3} , 3×10^{-3} , 1×10^{-2} , and 3×10^{-2} (cm/sec). Values of $\sigma = 1.5, 2.0$, and 2.5 were employed. T/H was specified as 1, 2, 5, 10, 50, 100, 500, $\infty \approx 10^{10}$ (sec/cm). Figures 14 through 37 show f_L plotted as a function of r_{50} , $\rho = 1$. There are four graphs on each figure, one for each γH . There are three sets of figures, one for $\sigma = 1.5, 2.0$, and 2.5 and eight figures in each set, one for each T/H . There are several ways to present the data shown in Figures 14 through 37. One method involves showing the dependence of f_L vs γH at different T/H 's as a function of R 's; see Figures 38 through 44 for this dependence when $\sigma = 2$. Each curve in a figure is for a different R_{50} . Another technique is to plot f_L vs T/H . There are four different leakage values of γH and three σ 's for each γH . Figure 45 shows the relationship of f_L vs T/H when R_{50} equals 5 microns. All curves shown in Figures 38 through 45 can be obtained from the original curves in Figures 14 through 37.

V. DISCUSSION OF RESULTS

Solutions of Equations 20 and 21 can be used for reactor siting evaluations as well as for aerosol physics studies. In the latter case, the values of f_R allow one to determine the fractional decrease of concentration with time if stirred settling (no agglomeration and no leakage) occurs; see Figures 6 through 13. Once T_{50} and σ are chosen, the time, t/H , for the concentration to decrease to the specified ratio can be determined from the figures. If the solution is desired for a particle density which is not equal to 1 gm/cc, the value of t/H obtained for r_{50} and t/H is corrected by dividing t/H for $\rho = 1$ by the new density. The values of R_{50} , the median aerodynamic radius, are obtained by multiplying r_{50} by the square root of the particle density; then t/H can be read directly. Actual time, in sec, is obtained by multiplying t/H (sec/cm) by H (cm), where H is the height of the container of concern. If it is required to know the fraction remaining in suspension when leakage occurs, the value of f_R can be corrected to

$$f_A = f_R \times e^{-(Q/V)t} = f_R e^{-\gamma t} = f_R \times \exp\left[-\gamma H(t/H)\right] .$$

The correction is independent of size, and the magnitude of the correction factor is insignificant until the fraction C/C_0 becomes very small and t/H or t is large.

As mentioned, the initial log-normal distributions of UO_2 , PuO_2 , and Na_2O aerosols have a geometric standard deviation, σ , in the range of 2 to 2.5, which decreases, as growth occurs. The experimentally observed air concentration appears to decrease in the early times with a single half-life. This is due to the narrowing of the distribution and growth of the effective size by agglomeration during the observation period, and by removal due to wall and floor deposition. A value of σ near 2 is likely to give very conservative estimates of the airborne suspension which remains, at very long times. This is true because:

- 1) The extremes of the distribution which are assumed to exist by the integration limits of Equations 20 and 21 are nonexistent; i.e., there are finite sizes which can be fitted to a log-normal distribution, but their range of existence is not from 0 to ∞ , and

- 2) At high concentrations of $>1 \mu\text{gm/cc}$, Brownian motion scavenges the small particles; and large particles settle, causing further narrowing of the distribution.

In addition to the selection of r_{50} , one has to estimate the material density and particle distribution of the released aerosol. When two or more materials of different density agglomerate to form a single aggregate, there are many possible combinations of density which individual aggregate particles may have. The ideal average density of the material which eventually settles is:

$$\bar{\rho} = \frac{C_R}{\frac{C_1}{\rho_1} + \frac{C_2}{\rho_2} + \frac{C_3}{\rho_3} + \frac{C_n}{\rho_n}},$$

where:

$C_R = \sum_i^n C_i$ and ρ_i' is the effective density of each released material. ρ_i' may be much less than the true density, ρ_i .

The effective r_{50} can be computed as:

$$(r_1)^3 + (r_2)^3 + \dots (r_n)^3 = (r_{50})^3,$$

where:

$r_n \sim f(C_R)_i^b$, and each r may have a different relationship to C_{Ri} as defined by the exponent, b . Again, r_{50} may have a different relationship to C_R in vessels with heights differing from that of Atomics International's LTC.

Equation 21 describes the relationship that is desired by the safety analyst; i.e.: the fraction of the released mass that leaks from the primary containment vessel when agglomeration occurs. At a given leak rate $\gamma H = 10^{-3} \text{ cm/sec}$, the leak fraction, $f_L(\infty)$ vs r_{50}^2 can be approximated as shown in Table 1, also see Figures 22 and 30 to obtain the approximations appearing in this table. At a constant σ , the leak fraction is inversely proportional to the radius, to a power. If normalized to $r = 1$ and $\rho = 1$, one observes that $f_L(\infty) \approx 66\gamma H \sigma^{1.23}$ and if the power of r is rounded off to r^2 , the approximate relations in Table 1 can be written as:

TABLE 1
APPROXIMATION OF LEAKAGE
FRACTION

Standard Deviation (σ)	Leakage $f_L(\infty)$	
	$\gamma H = 10^{-3}$ (cm/sec)	Normalized to γH
1.5	$\frac{0.1}{\rho' r_{50}^2}$	$\frac{100\gamma H}{\rho' r_{50}^2}$
2.0	$\frac{0.16}{\rho'(r_{50})^{1.94}}$	$\frac{160\gamma H}{\rho'(r_{50})^{1.94}}$
2.5	$\frac{0.19}{P'(r_{50})^{1.84}}$	$\frac{190\gamma H}{\rho'(r_{50})^{1.84}}$

$$f_L(\infty) \approx \frac{66\gamma H \sigma^{1.23}}{\rho r_{50}^2} \quad \dots (23)$$

This dependence is true only where $\sigma < 1.5$, $r > 1.54\mu$, and $\gamma H < 2 \times 10^{-3}$ cm/sec.

Thus, the fraction of the released mass which leaks depends on the particle size distribution characterized by r_{50} and σ . We may substitute the concentration equivalence of ρr_{50}^2 from Equations 11, 12, or 12a and rewrite Equation 23 in terms of concentration.

More accurate relations of f_L to r_{50} as a function of time should be read off the accompanying figures; however, Equation 23 can be used to show that the mass fraction which leaks increases as the geometric standard deviation, σ , the leak rate, γ , and the height, H , of the containment vessel increase, and then decreases as the released concentration, C_R or R_{50} , increases. The released concentration, of course, determines the value of R_{50} for a given geometry.

If agglomeration of all released materials and fission products are assumed to occur: then the fraction leaked, f_L , can be defined in terms of any specific material. The fraction, f_L of the original mass of material in the primary vault 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 presented by f_P , and f_{FP} , respectively. Thus, $M_R \cdot f_L \cdot f_P = \text{total mass of PuO}_2$, and $M_R \cdot f_L \cdot f_{FP} = \text{total mass of fission products which have leaked to the outer containment}$. 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 FP's to the total material released is computable from the fission products inventory of the irradiated fuel.

The activity in curies of a specific fission product isotope which has leaked is

$$f_L \cdot f_{FP} \cdot f_i \cdot f_n \cdot S \quad ,$$

where:

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

f_{FP} = fraction, a constant, of mass due to vaporized f_p's,

f_i = fraction of mass, f_p , due to species i ; f_i varies with time as isotopes of that species decay,

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

S = specific activity of isotope, n .

Since

$$f_{FP} \cdot f_i \cdot f_n \cdot M_R = M_n \quad ,$$

the mass of the isotope, n , produced at zero time;

$$f_L \cdot M_n = M_{Ln} \quad ,$$

the mass of isotope, n , which has leaked.

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 primary vaults; thus,

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

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 $n_z(\text{FP})$; and N_c , the total number of curies of a group of fission products ($z = \text{constant}$) or of an individual fission product, $n_z(\text{FP})$.

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.

The approximation shown in Equation 23 for $f_L(\infty)$ can be used to estimate the concentration of material in the secondary containment vessel, V_2 . Let the mass concentration of the material of concern be equal to

$$C_a = \frac{M_a}{V_1} \quad .$$

Then

$$f_a = \frac{M_a}{M_R} \quad ,$$

and

$$f_L = \frac{M_L}{M_R} = \frac{M_{La}}{M_a}$$

From Equation 23, we obtain, for the U-Fe oxide aerosols of Equation 11,

$$\frac{M_{La}}{V_2} \approx \frac{66\gamma H \sigma^{1.23} M_a}{4.4 C_R^{0.6} V_2} \approx \frac{66\gamma H \sigma^{1.23} f_a \cdot C_R^{0.4} V_1}{4.4 V_2} \quad ,$$

where $C_R = M_R/V_1$.

Thus, the leaked concentration of any specific material in the secondary vessel is a function of the total released mass concentration in the primary vaults, and the lower the released concentration, C_R , the lower the concentration in V_2 .

The leaked fraction, $F_L(\infty)$, at $t = \infty$ refers to the total leaked mass. The time to leak the total amount decreases as γH and r_{50} increase and as σ decreases. The effect of the particle size distribution on the amount of leakage vs time can be shown if one investigates the time for one-half of $f_L(\infty)$ to occur. Figures 47 through 50 show how T_{50L}/H for one-half $f_L(\infty)$ varies with r_{50} . Each figure represents a given leak fraction, γH . There are three curves on each figure, each one representing a different value of σ . In Figure 47, where $\gamma H = 10^{-3}$, T_{50L}/H can be approximately related to r_{50} as shown below for the linear portion of the curve.

Thus, the time for one-half of the mass to leak increases as H and σ increase, and decreases as r_{50} and the density increase.

<u>a</u>	$\frac{T_{50L}/H}{(\text{sec/cm})}$
1.5	$70/\rho(r_{50})^{1.8}$
2.0	$170/\rho(r_{50})^{1.74}$
2.5	$200/\rho(r_{50})^{1.2}$

VI. DETERMINATION OF THE EFFECTS OF AGGLOMERATIONS ON THE SITE RADIOLOGICAL SOURCE TERM

A. ILLUSTRATIVE EXAMPLE

A typical LMFBR DBA is postulated, and the models in this report are utilized to determine f_R , f_A , f_L , f_{PuO_2} , and M_{LPuO_2}/V_2 . The following geometry and accident conditions are assumed.

$$V_1 = 400,000 \text{ ft}^3 = 1.13 \times 10^{10} \text{ cc} \quad ,$$

$$V_2 = 2 \times 10^6 \text{ ft}^3 = 5.65 \times 10^{10} \text{ cc} \quad ,$$

$$M_R = 1680 \text{ kg} \quad ,$$

$$M_R(PuO_2) = 252 \text{ kg} \quad ,$$

$$M_R(UO_2) = 1008 \text{ kg} \quad ,$$

$$M_R(Na_2O) = 420 \text{ kg}$$

For a typical LMFBR, 1260 kg of PuO_2 - UO_2 represents $\sim 16\%$ of the core which is assumed to vaporize, oxidize and become airborne as an aerosol; some sodium is vaporized and oxidized simultaneously. The additional conditions are:

$$\gamma H = 1 \times 10^{-3} \text{ cm/sec} \quad ,$$

$$H = 920 \text{ cm} = 30 \text{ ft} \quad ,$$

$$\gamma = 0.095/\text{day} = 1.08 \times 10^{-6} \text{ sec}^{-1} \text{ for 1 day, and}$$

$$y = 0 \text{ after one day.}$$

For the purpose of this discussion, γ is assumed to remain a constant throughout the time period of one day. Actually, the leak rate fraction, y , varies with the pressure differential, ΔP , between V_1 and V_2 . As the temperature in V_1

decreases, $\mathbf{AP}$ and γ decrease. Under these circumstances, an average γ and $\mathbf{AP}$ can be selected for the time period of concern.

In the present analysis, it is assumed that the relation between the released mass concentration, C_R , and the mean size, r_{50} , to which the distribution agglomerates is the same as those of Equations 11 and 12. These computations are shown in Table 2, where

$$(\bar{r}_{50})^3 = (113)^{0.9} + (37.4)^{1.2} = (5.3\mu)^3,$$

and

$$\bar{\rho} = \frac{150}{\frac{113}{4.4} \times \frac{37.4}{2.27}} = 2.8,$$

$$\sqrt{\bar{\rho}}r_{50} = 8.8 = \bar{R}_{50}.$$

TABLE 2
RESULTS OF COMPUTED PARAMETRIC RELATIONSHIPS

Parameter	PuO ₂ t UO ₂	Na ₂ O	Mixture
M_{R_i} (kg)	1260	420	1680
C_{R_i} (μ gm/cm ³)	113	37.4	150
r_{50} (microns)	$(113)^{0.3}$	$(37.4)^{0.4}$	r_{50} (see above)
r_{50} (microns)	4.1	4.25	5.2
ρ (gm/cm ³)	4.4	2.27	ρ (see above)
$\sqrt{\bar{\rho}}r_{50} = R_{50}$	8.6	6.4	8.8 (see above)

As previously mentioned, the choice of σ is left up to the safety analyst; but it should be remembered that σ narrows as agglomeration occurs. In this example, we choose to let $\sigma = 2.0$ after the growth to R_{50} . We now have the information necessary to determine f_R , f_A and f_L vs time. By using $\sqrt{\bar{\rho}}r_{50} = R_{50}$ in the accompanying figures, the values of t/H which are read off the curves do not have to be corrected for density. Table 3 shows the calculations of f_R , f_A , f_L , and $f_L M_R(\text{PuO}_2)$.

TABLE 3
AIRBORNE FRACTION IN PRIMARY VAULT
($C_R = 150 \mu\text{gm/cc}$)

f_R	t/H at $R_{50} = 8.8$	$\gamma H(t/H)$	$e^{-\gamma t}$	f_A
0.9	0.052	5.2×10^{-5}	1	0.9
0.8	0.1	1.0×10^{-4}	1	0.8
0.5	0.65	6.5×10^{-4}	1	0.5
0.2	2.9	2.9×10^{-3}	0.997	0.1994
0.1	6.0	6×10^{-3}	0.994	0.0994
10^{-2}	33.0	0.033	0.967	0.967×10^{-2}
10^{-3}	110	0.11	0.896	0.896×10^{-3}
10^{-4}	280	0.28	0.756	0.756×10^{-4}

A reasonable time must be allowed for the growth to R_{50} from the initial distribution. As mentioned in Section III, the time to reach the maximum size is a function of the concentration. It should be renoted that R_{50} is not as large as R_{max} , and that R_{max} is a function of the height of the vessel. It is thus conservative to assume that R_{50} based on the LTC model is made instantaneously, and therefore the computations of f_L , as presented, are conservative.

Table 3 shows the computational procedure for calculating the airborne fraction remaining in the primary vault. Columns 4 and 5 of Table 4 show the mass leaked and the concentration in the outer containment vessel as a function of time.

After about 1 day, the concentration can be assumed to be $0.01 \mu\text{gm/cc}$. If the leak rate from V_2 into the environ is assumed to be 0.25%/day, then 1.26 gm PuO_2 leaks per day.

For a typical LMFBR Pu fuel isotope mixture and the worst condition meteorologically, the leakage of 1.26 gm in one day is equivalent to a (soluble PuO_2) bone dose of 58 rem at 10^4 ft. The pressure in the secondary vessel will not rise to values much above 1 psig and will not remain at these values much more than 1 day due to heat losses, Thus, for a vessel designed to leak 0.5%/day at

10 to 20 psig, the leaked PuO_2 would be nearer to 1/5 or 1/10 of 1.26 gm per day at a pressure difference equivalent to 1 psig, with a corresponding decrease in the dose rate, to 12 to 6 rem at 10^4 ft.

TABLE 4
RELEASED MASS TO OUTER CONTAINMENT VESSEL

t/H (sec/cm)	f_L at $R_{50} = 8.8$ and $\gamma H = 10^{-3}$ cm/sec ⁻¹	$t/H \times 920$ (sec)	$f_L \times M_R(\text{PuO}_2)$ (gm)	Concentration in Outer Containment ($\mu\text{gm/cc}$)
1	5.4×10^{-4}	920	136	2.4×10^{-3}
2	8.1×10^{-4}	1,820	201	3.6×10^{-3}
5	1.2×10^{-3}	4,600	300	5.3×10^{-3}
7.8	2 hr	7,200		
10	1.5×10^{-3}	9,200	380	6.7×10^{-3}
50	1.9×10^{-3}	46,000	480	8.5×10^{-3}
93.7	1 day	86,400		
100	2.0×10^{-3}	92,000	504	9×10^{-3}
500	2.0×10^{-3}	460,000	504	9×10^{-3}

B. CONSERVATISM OF THE ASSUMPTIONS IN THE ANALYSIS

There are several conservative assumptions inherent in this study which should be pointed out in order to show that the arguments listed above do not underestimate the potential hazard. These assumptions are listed below.

- 1) It is assumed that the particulates which are formed will leak through the primary vault leak paths in the same manner as a gas. No reduction has been made of particulate leakage due to impaction and fallout in the various assumed leak paths.
- 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. No reduction was made in concentration due to settling in the outer containment. This can be estimated from the knowledge of the particle sizes which have leaked from the hot cell. The particle sizes leaking were not computed in the current study.

- 3) All calculations assume that the leak rate is a constant throughout the time of interest. The leak rate is actually a function of temperature and pressure. The variations of these parameters with time can be computed from a thermal analysis of the energy released to the primary vaults and the resulting decrease of temperature and pressure due to the heat losses used to obtain the true leaked fraction.
- 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 primary vaults. When the leak fraction from the primary vaults is known as a function of time, see 3) above, the pressure rise in the secondary containment can be used to determine the leak fraction to the environment. Typically, the leak rate due to overpressure at 2 hr is considerably below that of the design pressure and effectively reduces the mass escaping to the environs.
- 5) Still another degree of conservatism is involved when one assumes that R_{50} is a function of the released mass concentration, C_R , and that a log-normal distribution exists with a constant σ . This comes about in the following manner.

The present model assumes that the measured T_{50} determines the effective size. The model allows the user to choose σ , R_{50} . Thus, no further growth of the particles which are airborne is assumed. It is observed experimentally, however, that growth continues for some time after the initial half-time occurs. The actual time that the growth continues after T_{50} , depends on the initial concentration; i.e., growth continues until fallout, plateout, etc. reduce the initial concentration to a low level. When the height of the laboratory test chamber is used to normalize the relationship of the initial half-time to the released concentration, it is assumed that growth is independent of height. Actually, taller enclosures allow more time for agglomeration.

Thus, the relation of R_{50} to C_R is conservative when based on the small test chamber results and R_{50} is less than $R_{\max}$.

VII. SUMMARY AND CONCLUSIONS

This report shows that the leaked fraction of the radioactive materials released during a LMFBR-DBA may be considerably reduced due to agglomeration and settling of the aerosols. A computational procedure is given for studying the reduction factor of the radiological source term as a function of the aerosol particle size distribution. An empirical relation is derived from experiments performed at Atomics International in a small test chamber, which relates the mass concentration, in partially sealed containers, to the aerosol particle size distribution which agglomeration produces. When the particle size distribution, the leak rate, and the height of the enclosure are specified, the leaked fraction at any time can be read from the figures presented in this report.

If a functional dependence which is different from that recommended in this report is used between the concentration and size, the computed relations of leak and size are still applicable. The particulates of each material are assumed to agglomerate into aggregates containing a mixture of all of the airborne species, and thus the leaked fraction of the total mass is indicative of that of any other species.

The model developed in this report is recommended as a tentative solution to a very complicated problem which is being studied at many laboratories throughout the world. The model is thought to be both conservative, physically realistic, and usable for practical evaluations of LMFBR radiological safety until a more rigorous treatment can be developed.

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PLOT CHARACTERS FOR
Figures 6 through 13

STIRRED SETTLING CGCE

PLOT CHARACTERS FOR SIGMA = 1.0 WILL BE O

PLOT CHARACTERS FOR SIGMA = 1.5 WILL BE +

PLOT CHARACTERS FOR SIGMA = 2.0 WILL BE C

PLOT CHARACTERS FOR SIGMA = 2.5 WILL BE X

Figure 6.

Figure 7.

Figure 8.

Figure 9.

Figure 10.

Figure 11.

Figure 12.

Figure 13.

PLOT CHARACTERS FOR
Figures 14 through 37

STIRRED SETTLING CODE VERSION 3

PLOT CHARACTERS FOR GAMMACH = 1.00000E-03 WILL BE 0

PLOT CHARACTERS FOR GAMMACH = 3.00000E-03 WILL BE •

PLOT CHARACTERS FOR GAMMACH = 1.00000E-02 WILL BE C

PLOT CHARACTERS FOR GAMMA*H = 3.00000E-02 WILL BE X

Figure 14.

Figure 15.

Figure 16.

Figure 17.

Figure 18

Figure 19.

Figure 20.

Figure 21.

Figure 22.

Figure 23.

Figure 24.

Figure 25.

Figure 26.

Figure 27.

Figure 28,

Figure 29.

Figure 30.

Figure 31.

Figure 32

Figure 33.

Figure 34.

Figure 35.

Figure 36.

Figure 37.

PLOT CHARACTERS FOR
Figures 38 through 45

STIRRED SETTLING CODE VERSION 3

PLOT CHARACTERS FOR R5D = 0.5 WILL BE O

PLOT CHARACTERS FOR R5G = 1.0 WILL BE +

PLOT CHARACTERS FOR R5D = 2.0 WILL BE C

PLOT CHARACTERS FOR R5D = 3.0 WILL BE X

PLOT CHARACTERS FOR R5G = 5.0 WILL BE H

PLOT CHARACTERS FOR R5D = 10.0 WILL BE *

PLOT CHARACTERS FOR R5D = 15.0 WILL BE U

PLOT CHARACTERS FOR R5G = 25.0 WILL BE E

Figure 38.

Figure 39.

Figure 40.

Figure 41.

Figure 42.

Figure 43.

Figure 44.

Figure 45.

PLOT CHARACTERS FOR

Figure 46

STIRREC SETTLING CODE VERSION 3

PLOT CHARACTERS FOR GAMMA*H = 1.0E-03 SIGMA= 1.5 WILL BE O

PLOT CHARACTERS FOR GAMMA*H = 1.0E-03 SIGMA= 2.0 WILL BE +

PLOT CHARACTERS FOR GAMMA*H = 1.0E-03 SIGMA= 2.5 WILL BE C

PLOT CHARACTERS FOR GAMMA*H = 3.0E-03 SIGMA= 1.5 WILL BE X

PLOT CHARACTERS FOR GAMMA*H = 3.0E-03 SIGMA= 2.0 WILL BE H

PLOT CHARACTERS FOR GAMMA*H = 3.0E-03 SIGMA= 2.5 WILL BE *

PLOT CHARACTERS FOR GAMMA*H = 1.0E-02 SIGMA= 1.5 WILL BE U

PLOT CHARACTERS FOR GAMMA*H = 1.0E-02 SIGMA= 2.0 WILL BE E

PLOT CHARACTERS FOR GAMMA*H = 1.0E-02 SIGMA= 2.5 WILL BE .

PLOT CHARACTERS FOR GAMMA*H = 3.0E-02 SIGMA= 1.5 WILL BE D

PLOT CHARACTERS FOR GAMMA*H = 3.0E-02 SIGMA= 2.0 WILL BE \$

PLOT CHARACTERS FOR GAMMA*H = 3.0E-02 SIGMA= 2.5 WILL BE M

Figure 46.

PLOT CHARACTERS FOR
Figures 47 through 50

STIRRED SETTLING CODE VERSION 3

PLOT CHARACTERS FOR $\Sigma = 1.5$ WILL BE •

PLOT CHARACTERS FOR $\Sigma = 2.0$ WILL BE C

PLOT CHARACTERS FOR $\Sigma = 2.5$ WILL BE X

Figure 47.

Figure 48.

Figure 49.

Figure 50.