AN EVALUATION OF THE BEHAVIOR
OF AEROSOLS PRODUCED BY
THE LMFBR DBA

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CONTENTS

	Page
Acknowledgement	4
Abstract	5
I. Introduction	6
II. Experimental Data and Theoretical Models	8
III. Summary and Conclusions	15
References	16

TABLE

1. Comparison of Available Experimental Data to DBA Conditions	9
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FIGURES

1. Initial Concentration vs Initial Half-Time	10
2. Aerodynamic Equivalent Diameter (AED) vs Concentration	13
3. Time After End-of-Burning/Height of Chamber	14

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ABSTRACT

The existing experimental data on UO_2 and PuO_2 aerosols are evaluated and an empirical model of the behavior of aerosols produced in the primary containment by the LMFBR DBA is developed. This model is based on the well known "stirred settling" law for aerosols. It permits a conservative, but physically realistic, estimate of the specific reduction factor for the aerosol concentration in the primary containment for any given plant due to agglomeration and subsequent fallout. Such an estimate should provide an adequate and useful interim guide for both LMFBR designers and DRL staff.

I. INTRODUCTION

The typical design basis accident (DBA) of an oxide fueled LMFBR⁽¹⁾ results in the vaporization of 3 to 20% of the core (PuO_2 and UO_2) and the rapid ejection of this mass through the upper sodium pool and into the cover gas space above. The excess pressure accompanying the DBA causes the aerosols to vent into the primary containment volume. The predicted temperature of the vaporized core materials is initially $\sim 6000^\circ\text{C}$. These materials will rapidly condense to form branched chain aggregates (as shown by experiments reported in References 2, 3, and 4). Any fission products or vaporized sodium which is present may condense on the branched-chain structures and form even more complex aerosol particulates. The predicted initial concentration of airborne particulates in the primary containment will be in the range of 20 to 200 $\mu\text{gm/cc}$.

To estimate the specific reduction factors for the accident-produced aerosols in a given plant, we have evaluated the existing experimental data on UO_2 and PuO_2 aerosols and have developed an empirical model of the subsequent behavior of these high concentration aerosols produced in the primary containment by the LMFBR DBA. It will be shown that the initial concentration in the primary system is rapidly reduced by naturally occurring agglomeration and fallout so that large specific reduction factors are obtained (depending upon initial concentration).

The extrapolation of the result of aerosol experiments done on a laboratory scale ($40 \rightarrow 2000 \text{ ft}^3$ volume) to containment volumes of 4×10^5 to $1.5 \times 10^6 \text{ ft}^3$ cannot be done directly but requires consideration of the theoretical behavior of aerosols since the aerosol clearance characteristics are affected by the released concentrations, the height and the surface to volume ratio of the experimental chamber, and the aerodynamic characteristics of the major aerosol components.

In the case of the leakage of aerosols from one vessel to another or to the atmosphere, the subsequent particle size distribution depends on the aerosol size distribution and concentration in the primary vessel and the time during which the leakage takes place. As settling occurs, the distribution which remains or leaks changes, eventually reaching a sufficiently small concentration where further growth does not occur.

These features are explainable from the laws of aerosol physics which have been well demonstrated over the years. In particular, at high concentrations such as would occur in the postulated DBA, the small particle sizes quickly disappear as they agglomerate into successively larger particles which fall out. Under these conditions, it is impossible for fine aerosols to avoid agglomeration when the initial concentration is high. This process eliminates any extremely long-lived aerosol.

II. EXPERIMENTAL DATA AND THEORETICAL MODELS

Table 1 compares the available experimental aerosol data with the aerosol characteristics of the LMFBR DBA. Figure 1 illustrates how the experimental data for the ratio of T_{50_i} (the time for the initial concentration to decrease by 1/2) to H (the vessel height) are related to the initial concentration, C_o . Evaluated data* on vaporized UO_2 and PuO_2 aerosols are presented from three different laboratories.⁽²⁻⁴⁾

The data of Figure 1 can be represented by:

$$\frac{T_{50_i}}{H} = a C_o^{-p} \quad , \quad \dots (1)$$

where

C_o = initial concentration (μ gm/cc)

H = chamber height (cm)

T_{50_i} = initial half-time (sec); i. e., $t = T_{50_i}$ when $C = C_o/2$.

a and p = constants determined by the data of Figure 1.

A monosize model[†] with an effective mean radius, r , is assumed and occurrence of stirred settling is also assumed. The following equation⁽⁶⁾ may then be written for the mass concentration, C ,

$$\frac{dC}{dt} = - \left(\frac{v_s}{H} \right) (C) \quad . \quad \dots (2)$$

For fallout, the settling velocity, v_s , is given by:⁽⁵⁾

$$v_s = 0.012 r^2 \rho \quad , \quad \dots (3)$$

*The data presented in Figure 1 were abstracted from BNL⁽⁴⁾ and ORNL⁽³⁾ reports but in a different form from that of the original publication. In plotting these data for Figure 1, it is assumed that the initial rapid decrease was due to local effects and thus does not represent true settling of some initial distribution. Only the long tail component of these data has been used to extrapolate to the C_o used in Figure 1 (a conservative assumption). In plotting the BNL data on $PuO_2 + Na$ aerosols, the sodium concentration has been added to the plutonium concentration.

†Even though it is obviously impossible for an agglomerating aerosol to remain monodispersed, the weak dependence of the agglomeration on actual particle size distribution justifies the use of a monosize model.

TABLE 1
COMPARISON OF AVAILABLE EXPERIMENTAL DATA TO DBA CONDITIONS

Parameter	BNL (~3200°C)	ORNL (~4000°C)	AI (~4000°C)	LMFBR DBA (~6000°C)
C_o (μ gm/cc)	$10^{-5} - 10^{-1}$	$5 \times 10^{-3} - 0.6$	0.06 - 20	20 - 200
Initial MMD (μ)	0.6 - 0.8	0.2	0.7 - 1.0	0.2 - 1.0
Geometric Standard Deviation of Initial Distribution (σ)	1.5 - 2	1.5 - 2.0	1.5 - 2.5	1.5 - 2.5
Aerosol Production Method	vaporization indirect heating (inert gas)	vaporization plasma torch (air)	vaporization electric arc (air)	nuclear excursion
Chamber Volume (ft^3)	28	1072	40	400,000
Height (ft)	3-1/4	15	6	30
Material	UO ₂ , Na, PuO ₂	UO ₂ , SS, FP	Na, UO ₂ , SS, FP	PuO ₂ , UO ₂ , FP, SS
Sampling	min-hr at 7 cc/ min, N ₂ , A	air	5 sec-1 hr at 1 l/min air	
Measurement	size distribution and air concentra- tion vs time	air concentra- tion vs time and size distribution	concentration in air, mass settling, mass plating, and size distribu- tion vs time	
Reference	(4)	(3)	(2)	(1)

where

v_s = settling velocity (cm/sec)

ρ = effective density of aerosol.

For stirred settling, Equation 2 shows:

$$T_{50} = \frac{0.693H}{v_s} \quad \dots (4)$$

Therefore from Equations 1, 2, 3, and 4,

$$T_{50_i} \propto \frac{1}{C_o P} \propto \frac{1}{r^2}, \text{ or } r \propto C_o^{1/2} P^{1/2}.$$

Figure 1. Initial Concentration vs Initial Half-Time

This suggests that r may be a function of C . It is therefore assumed:

$$r = kC^n, \quad \dots(5)$$

where

k = a constant to be determined

n = a constant to be determined

C = concentration (μ gm/cc)

r = mean effective particle radius (μ meters).

The preceding equations may be combined and integrated and a relationship deduced to be compared with Equation 1,

$$\frac{dC}{dt} = -\frac{(0.012)\rho k^2}{H}(C)^{1+2n}.$$

Integrating with $C = C_o$ at $t = 0$,

$$\frac{1}{C^{2n}} - \frac{1}{C_o^{2n}} = \frac{(0.012)(\rho k^2)(2nt)}{H} \quad \dots(6)$$

Let $C = 1/2C_o$ at $t = T_{50_i}$, then,

$$T_{50_i} = \left[\frac{(2^{2n} - 1)(H)}{(0.012)(\rho k^2) 2n} \right] (C_o)^{-2n} \quad \dots(7)$$

Identifying Equation 7 with Equation 1,

$$n = \frac{p}{2} \quad \dots(8)$$

and

$$k = \frac{1}{\sqrt{\rho}} \left[\frac{2^p - 1}{(0.012 p) a} \right]^{1/2} \quad \dots(9)$$

If $a = 40/3$ and $p = 0.6$ from Curve A*, Figure 1, then $n = 0.3$ and $k = 2.34/\sqrt{\rho}$, so that Equation 5 becomes:

$$r = \frac{2.34}{\sqrt{\rho}} (C)^{0.3} \quad \dots(10)$$

*The floor settling data are used in preference to the airborne data since the latter include wall plating effects which occur in the small experimental chambers. The use of the floor settling data is conservative as the predicted half-times are larger than for the airborne data.

If "a" is reduced in value by a factor of 2 (approximately to the position of Curve B, the airborne particle half-time vs C_o given in Figure 1), the value for k increases by a factor of $\sqrt{2}$.

If p is increased to 0.7, the effect on k is not great; in that case:

$$k = \frac{2.36}{\sqrt{\rho}} \quad .$$

Equation 10 is compared to AI laboratory experiments* on UO_2 aerosols in Figure 2. It is seen that the aerodynamic equivalent diameter, AED, given by Equation 10 is less by about a factor of 2 than those obtained experimentally for the same concentration, C. Thus, the present model predicts an airborne concentration which will persist for longer times than will actually be the case.

Figure 3 was prepared by considering Equation 6 and shows the decrease with time of several initial concentrations in a containment building. The abscissa is plotted in units of t/H in sec/cm. (To compute the actual elapsed time for the reduction of the concentration to a given value for a building of a given height, H, the abscissa values were multiplied by H in cm.) The initial rapid decrease noted for Curve A is a direct result of the relationship shown in Figure 1 and Equation 1 and verified by tests at AI performed in a 6-ft high vessel.⁽²⁾

Curve B was prepared from a simultaneous solution⁽⁶⁾ of the classical equations for Brownian agglomeration. (Smolochowski, Müller, Goldman) and the equations for particle agglomeration due to gravitation (Saffman and Turner).

Again it can be seen in Figure 3 that the specific reduction factors predicted by the present model are smaller than the results obtained from both a more exact theory based on first principles and from experiments on UO_2 aerosols performed at AI.

*The experimental points⁽²⁾ were obtained by measuring dM_f/dt in gm/cm²-sec, the rate of mass deposition of UO_2 aerosol on the floor as a function of C, the airborne concentration. These quantities are related by:

$$\frac{dM_f}{dt} = v_s (C)$$

From Equation 3, $v_s = 0.012\rho r^2$, or

$$AED = 2r \cdot \sqrt{\rho} = \sqrt{\frac{v_s}{0.003}} = \sqrt{\left(\frac{1}{0.003}\right) \left(\frac{dM_f}{dt}\right) \left(\frac{1}{C}\right)}$$

Figure 2. Aerodynamic Equivalent Diameter (AED) vs Concentration

Equation 6 can be modified to account for leakage from the vessel and wall plating effects. By this technique the fraction of the initial airborne mass which leaks can be determined and the atmospheric radiological source term can be derived.⁽⁷⁾

Figure 3. Time After End-of-Burning/Height of Chamber

III. SUMMARY AND CONCLUSIONS

Figure 1 and Equation 1 show the effect of initial concentration, C_o , and height, H , on the initial fallout half-time, T_{50i} , and the mass median radius, r , to which the initial distribution grows. Figure 3 shows that the rate of change of concentration is proportional to the initial concentration.

Agglomeration and settling are still occurring at initial concentrations of 10^{-4} $\mu\text{gm/cc}$. Agglomeration does not directly decrease the mass concentration of aerosols, it only decreases the number of particles per cm^3 and increases the effective size. In the 6-ft high experimental chambers, the mass concentrations decrease by both fallout and wall plating mechanisms, and of course the larger particles settle faster and contain the greatest mass per particle.

In the present model, the effective particle radius, r , is directly related to the mass concentration, C , by Equation 10.

As more mass is released into a given volume, the particle concentration increases and r increases to a maximum in proportion to the released mass concentration, C_o , from some initial effective size, r_o . As the particles settle, the mass decreases and the effective radius decreases since the larger particles settle first.

The model presented herein is considered to be conservative with respect to actual LMFBR accidents since:

- 1) It can be shown from Na_2O aerosol experiments⁽⁹⁾ that the t/H dependence of the present model is conservative when applied to taller vessels.
- 2) No growth of the aerosol is assumed after the initial growth.
- 3) No removal of the aerosol is assumed due to wall plating.

Because of these assumptions, we believe that the present model provides a conservative analysis of aerosol behavior following the LMFBR DBA. It can be used as an interim analysis technique while more exact characterization of the details of the problem is being developed.

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