CONSIDERATIONS IN THE BEHAVIOR OF
AEROSOLS IN GLOVEBOXES. I. MODELS

by

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ABSTRACT

Increasing requirements for minimizing aerosol release from gloveboxes and minimizing personnel radiation in the processing and recycling of reactor grade plutonium necessitate a knowledge of the behavior of aerosols in such enclosures.

This paper presents the results of a literature survey concerning aerosols in gloveboxes. The results of some theoretical plutonium aerosol models in simulating glovebox conditions are reported.

INTRODUCTION

Physical barriers and directional airflow have historically been used to confine radiotoxic plutonium aerosols generated by processing and handling plutonium. In the past, the quantities of penetrating radiation associated with plutonium have not been a serious problem. With the advent of processing of commercial reactor grade plutonium and the potential for recycle, the ALARA (As Low As Reasonably Achievable) philosophy dictates minimizing personnel radiation from handling and processing as well as minimizing airborne release.

Up to now, the prime considerations in the design of gloveboxes have been confining the aerosols within gloveboxes and ease of operations, without due regard for what effect the design may have on the behavior of the

aerosol within the glovebox. It is believed the potential dose from handling and processing, and the potential airborne release from leaks can be minimized if the factors controlling the behavior of aerosols in gloveboxes are understood and utilized.

A program of studies has been initiated by the Engineering and Analysis Section of the Hanford Engineering Development Laboratory to provide such information. The program is divided into three parts: 1) modeling of the behavior of confined aerosols, 2) experimental simulation of aerosol behavior within a glovebox, and 3) measurement of aerosol parameters in an actual process glovebox. This document addresses item 1.

DISCUSSION

I. Literature Search

Very little information on the behavior of aerosols in gloveboxes was uncovered. Literature on gloveboxes was primarily directed towards design and operational considerations to assure the integrity of the physical barriers and directional flow, plus ease of operations. A comprehensive review of such considerations is given in Chapter 7 of Burchsted, Kahn, and Fuller's document.¹

There are a number of studies describing various models to simulate the behavior of aerosols within confined volumes. A computer code, HAA-3 (Heterogeneous Aerosol Agglomeration), simulates the changes in size distribution and concentration of a well mixed, log-normally distributed aerosol in a cylinder due to agglomeration, settling, and plating under static conditions.² PARADISEKO³ is a similar code, but it calculates aerosol behavior in 40 different particle size groups, instead of using a log-normal distribution. Results of HAA-3 and PARADISEKO compare favorably. A discussion of these and other similar codes and their applications was presented by Huang and Stitt⁴. A number of programs have been reported to simulate the velocity fields of gases or liquids in various flow arrangements. The Marker and Cell (MAC)⁵ and the Simplified MAC (SMAC)⁶ methods are well known computer codes which solve the Navier-Stokes equations and the continuity equation

in two dimensions for incompressible flow using a finite difference, iterative method. KACHINA⁷ is a code which computes two dimensional, two-phase flow in cylindrical systems by finite difference methods. YAQUI⁸ is a complex, variable mesh scheme which can calculate flow of individual particles in a two-dimensional cylindrical system without agglomeration. SOLA⁹ (Solution Algorithm) was developed from MAC and SMAC to offer a simplified version for persons inexperienced in numerical fluid dynamics. The SOLA code (available from the Argonne Code Center) doesn't contain the refined options built into MAC and SMAC codes. In addition to these published two-dimensional codes, a number of reports describing fluid and particle flow in three dimensions are in the literature.^{10,11,12} Unfortunately, these codes weren't listed in the articles and they don't appear to be available. In private communication, C. W. Hirt, L.A.S.L., has stated that a three dimensional version of SOLA is being documented and may be available in the future.

No particle moving computer code listings were found in the literature. However, a particle moving code, SOLA-PART, was obtained from C. W. Hirt. This code calculates particle movement by convection and diffusion in a uniform, velocity field, and settling due to gravity. SOLA-PART is currently set up for the two dimensions.

II. Model Applications

HAA-3. The program uses an approximation to the theoretical model of aerosol behavior, namely the size distribution function is log-normal and the parameters which characterize this distribution are a function of time.² All calculations are for spherical particles with a bulk density adjustment factor. The code assumes a uniform mixture at all times. As long as the size distribution is log-normal, the model can calculate changes in the aerosol due to particle generation, Brownian and gravitational agglomeration, settling, plating, and leakage removal mechanisms. When the size distribution no longer fits the log-normal criterion, the code switches to a stirred settling model (SSM) where settling with complete

mixing is the only depletion mechanism. The reader is referred to reference 2 for a detailed discussion of the HAA-3 theory.

The HAA-3 code was used to estimate the relative importance of agglomeration, which is not available in the SOLA-PART program. The following values were constant for all runs:

Initial Geometric Standard Deviation	2.0
Particle Density	11.50 g/cc
Density Modification Factor	0.25
Glovebox Height	4 ft
Glovebox Volume	128 cu ft
Temperature	300° K
Air Density	1.18×10^{-3} g/cc
Air Viscosity	1.8×10^{-4} poise
Width of Concentration Gradient	10^{-5} cm
Gravitational Agglomeration Efficiency	1.0

Variable input parameters were:

Plating	yes/no
Brownian Agglomeration	yes/no
Gravitational Agglomeration	yes/no
Initial Concentration, C_0	0.1/0.01 μ g/cc
Initial Mass Median Radius r_m	0.035/0.35 μ m

The designation for individual runs is GLVBX followed by two symbols randomly assigned by the computer. The assignment of variable inputs is Tabulated in Table I.

TABLE 1. Input Parameters for HAA-3 GLVBX RUNS

RUN	$r_m(\mu m)$		$C_o(\mu g/cc)$		PLATING	BROWN	GRAVIT.
	0.35	0.035	0.1	0.01		AGGL	AGGL
N9	X		X		Yes	Yes	Yes
ϕH	X		X		Yes	-	-
JD	X		X		-	-	-
D0	X		X		-	Yes	Yes
JB	X		X		-	Yes	
D4		X	X		-	Yes	-
JU		X	X		-	Yes	-
KA		X	X		-	-	-
WD	X			X	-	-	-
VS	X			X	-	Yes	Yes
WL		X		X	-	-	-
WR		X		X	-	Yes	-

An apparent artifact of the application of the HAA-3 to the conditions for this study was revealed during the course of the study. Under some conditions (runs N9 and ϕH), the airborne concentration of non-agglomerating aerosols was apparently decreasing faster than an agglomerating aerosol (see Fig. 1). The anomaly is an apparent effect of the influence assigned to plating in the programs. Runs N9 and ϕH used identical r_m and C_o , but agglomeration was included in N9. After 650 sec, the size distribution of N9 deviated from the log-normal and the program switched to the SSM which does not include plating. Two additional runs, D0 and JD, were run with the r_m and C_o identical to N9 and ϕH , but without the plating function. The calculated rate of change of the airborne concentration (C/C_o) is also plotted in Fig. 1 and further demonstrates the effect of the plating function.

The potential influence of plating under the conditions of this study was considered and was judged to be overstated by the HAA-3 program. Under the current flow rate (0.4 cfm), the postulated glovebox would not

FIGURE 1. Suspended Mass Concentration Versus Time from HAA-3 Computer Runs Comparing Effects of Plating and Agglomeration

approach the ideal well mixed, uniformly distributed aerosols assumed by the program. Aerosols would tend to concentrate near their point of generation and be carried slowly downstream. The plume would be spread by diffusion and mixing. Under these conditions, the concentration gradient at the wall would not approach the values calculated in the HAA-3 and would only exist over a limited fraction of the surface of the glovebox. Thus, it was felt the use of plating function in the HAA-3 would cause an unrealistically rapid depletion of the glovebox aerosols and its use was discontinued.

The effect of various other input parameters (r_m , C_0 , Brownian agglomeration, and gravitational agglomeration) on the aerosol depletion rate were assessed in the other computer runs. The results are plotted in Figs. 2 and 3. The results indicate:

1. The effect of gravitational agglomeration is negligible when compared to Brownian agglomeration at the particle radii (0.35 and 0.35 μm) and initial concentrations (0.1 and 0.01 $\mu\text{g/cc}$) used. Compare runs JB and D0 and runs D4 and JU.
2. Agglomeration does not have a significant effect on the aerosol mass depletion rate under the conditions used for approximately 10^4 sec. (Smaller particles and higher initial concentrations are most affected. See Fig. 2.)
3. At an initial mass median diameter (r_m) of 0.35 μm , the particle number density does not indicate a significant decrease for the first 10^2 sec, but shows a significant decline in 1 sec at a $r_m = 0.035$ μm (indicates particle agglomeration occurs rapidly at $r_m = 0.035$ μm , but particles do not grow to a size significantly influenced by settling for 10^4 sec). Compare runs D0 and D4 on Fig. 3.

SOLA and SOLA-PART. SOLA is a two-dimensional finite difference solution of the Navier-Stokes equations and the continuity equation. Details of the theory of SOLA are in reference 9.

SOLA-PART was originally set up to move particles in a uniform flow

FIGURE 2. Suspended Mass Concentration Versus Time from HAA-3 Computer Runs Without Plating

FIGURE 3. Particle Concentration Versus Time from HAA-3 Computer Runs Without Plating

field by convection, diffusion, and settling. It has been converted to use the flow field generated by SOLA. Limitations of SOLA-PART include:

1. Only one particle size and density are used in a run.
2. There are no means for particle agglomeration.
3. Only 2-dimensional solutions are currently available.

In SOLA-PART, the equations for particle movement are developed by the following scheme. Consider a single spherical particle shown in Figure 4. In the vertical direction, the force balance can be written as

$$(1) \quad F = ma = C_D \pi r^2 \rho_f \frac{V^2}{2} + \rho_p \frac{4}{3} \pi r^3 G$$

Where "a" is the net particle acceleration and the other symbols are described in Figure 4. This reduces to

$$(2) \quad \frac{\partial V_p}{\partial t} = a = G + \frac{3}{8} \frac{\rho_f C_D V^2}{\rho_p r}$$

where V_p equals particle velocity. For a particle Reynolds Number less than 1, C_D can be approximated by

$$(3) \quad C_D = 12 \frac{\mu_f}{r \rho_f V}$$

where μ_f equals fluid viscosity.

Substituting Equation (3) into (2) yields

$$(4) \quad \frac{\Delta V_p}{\Delta t} \cong \frac{\partial V_p}{\partial t} = G + \frac{9}{2} \frac{\mu_f V}{r^2 \rho_p}$$

To incorporate diffusion into the problem assume a "diffusion velocity" such that

$$(5) \quad V = V_f + V_d - V_p$$

where V_f is fluid velocity and V_d is "diffusion velocity". Equation (4) reduces to

- C_D = STOKES' DRAG COEFFICIENT
 G = GRAVITATIONAL ACCELERATION
 r = PARTICLE RADIUS
 ρ_p = PARTICLE DENSITY
 ρ_f = FLUID DENSITY
 V = VELOCITY OF FLUID RELATIVE TO PARTICLE

FIGURE 4. Vertical Force Balance on a Particle

$$(6) \quad \Delta V_p = G\Delta t + K\Delta t (V_f + V_D - V_p)$$

where $K = \frac{9}{2} \frac{\mu_f}{r_p^2 \rho_p}$

For small time increments, $V_p \approx V_{p(t+\Delta t)}$ on the right side of equation (6), and this reduces to

$$(7) \quad V_{p(t+\Delta t)} = \frac{V_{p(t)} + G\Delta t + K\Delta t (V_f + V_d)}{1 + K\Delta t}$$

This equation is used in both horizontal and vertical directions on each particle for each time step in SOLA-PART.

A more practical assumption would be $V_p \approx \frac{V_{p(t+\Delta t)} + V_{p(t)}}{2}$. The final equation would be

$$(8) \quad V_{p(t+\Delta t)} = \frac{V_{p(t)} + G\Delta t + K\Delta t \left(V_f + V_d - \frac{V_{p(t)}}{2} \right)}{1 + \frac{K\Delta t}{2}}$$

This equation should be incorporated into SOLA-PART.

The "diffusion velocity" is calculated in the manner described by Hotchkiss¹⁰.

The SOLA program was adapted to the BCS cyber system with appropriate matrix values for a 4' X 8' glovebox and designated SOLA-1. The output data are a graphic display of the velocity in each cell during various pre-selected cycles. The program also generates a card deck of velocities in each cell after approximately 30 cycles - a point after which it was determined by experimentation that no significant changes in velocity occur.

A case simulating a flow of 0.4 cfm air through the box has been performed and the results are shown graphically in Figure 5. The velocity

0.4000 CFM
210.0000 SECONDS

VELOCITY VECTORS

FIGURE 5. Velocity vector plot simulating 0.4 cfm flow through a two-dimensional glovebox

vector magnitudes are relative to the inlet velocity magnitude. In the real glovebox, the normal exit velocity is 1/4 of the nominal inlet velocity. (Gas is introduced via a single 8" X 8" HEPA filter and exhausted via four 8" X 8" HEPA filters.) Because SOLA gives only a 2-dimensional simulation of the flow field, the exit dimension is twice the inlet dimension and thus the exit velocity must be 1/2 the inlet velocity. The inlet velocity was reduced to 1/2 the actual velocity to compensate for this program limitation. The four obstacles shown (from left to right) are intended to represent equipment currently in use in PS-22 (the Blending Glovebox) are: 1) balance used to weigh oxides; 2) shaker and sieves; 3) "V" blender; and 4) ball mill.

Preliminary runs to determine the flow fields at 5 and 50 cfm have revealed some additional problems. Velocities in the cells are calculated and recalculated until a convergence criterion entered into the program is satisfied. Hundreds of iterations have failed to satisfy the convergence criteria at these flow rates. C. W. Hirt (LASL) has been consulted on the significance of the criteria. He suggested several changes in the program to accelerate convergence to an adequate solution. However, velocity data for 5 and 50 cfm have not been obtained.

SOLA-PART produced graphic output showing particle positions at specified times. Also, for each time increment the number of particles collected on floors, walls, filters, and obstacles is printed.

A graphic record of particle movement in a 0.4 cfm flow field is shown in Figure 6, 7, and 8. Initial conditions in all three cases are that 500 particles 0.1 μ m in diameter are released in a circle in the middle of the glovebox (in the cell just above obstacle #3). Densities of 2.5, 5.0¹ and 10 g/cc were used for SOLAPUT, SOLAPOV, and SOLAPPX, respectively.

CONCLUSIONS AND RECOMMENDATIONS

The three programs (HAA-3, SOLA, and SOLA-PART) appear to have some applicability to the understanding of the behavior of aerosols in gloveboxes.

FIGURE 6. 0.1 micron diameter particles in a 0.4 cfm simulated flow field. Particle density is 2.5 g/cc

FIGURE 7. 0.1 micron diameter particles in a 0.4 cfm simulated flow field. Particle density is 5.0 g/cc

FIGURE 8. 0.1 micron diameter particles in a 0.4 cfm simulated flow field. Particle density is 10.0 g/cc.

HAA-3 appears to be limited, but it is useful to indicate the relative importance of the agglomeration mechanism that is absent in SOLA-PART. The SOLA programs are versatile and show great promise.

Results from the HAA-3 program indicate particles of MMD of $0.7\text{ }\mu\text{m}$ at an initial concentration of $0.1\text{ }\mu\text{g/cc}$ do not agglomerate rapidly and the behavior of such particles may be adequately described by SOLA-PART. However, particles of MMD of $0.07\text{ }\mu\text{m}$ at initial concentrations of 0.01 and $0.1\text{ }\mu\text{g/cc}$ agglomerate rapidly and need compensatory assumptions (adjusted MMD and particle density) before SOLA-PART can be applied. HAA-3 also showed that gravitational agglomeration is insignificant compared to Brownian agglomeration.

SOLA has been used to develop a flow field in a simulated two dimensional glovebox. The flow rate is an approximation to the current flow rate (0.4 cfm) used in glovebox PS-22 in the Plutonium Fuels Lab. Velocity fields for 5 and 50 cfm have not been produced yet due to problems with convergence which are currently being worked on.

SOLA-PART has been converted to use the flow field produced by SOLA, and three cases for different density particles have been run. There appears to be little effect of changing density of particles while holding the flow constant at 0.4 cfm (simulated), and particle size constant ($0.1\text{ }\mu\text{m}$). SOLA-PART appears to be ready for more detailed study of different particle sizes and densities at simulated flow rates of 0.4 cfm .

Further work is needed to obtain velocity data simulating 5 and 50 cfm . When this problem is solved, conversion of the codes to three dimensions appears promising if experimental data tends to agree with the 2-dimensional results.

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