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REMOVAL OF RADIOACTIVE
PARTICLES BY SPRAYS

L. F. Parsly

OAK RIDGE NATIONAL LABORATORY
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Reactor Division

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CONTENTS

	<u>Page</u>
Foreword	v
Abstract	1
Introduction	1
Theory of Particle Capture by Spray Drops	2
Agglomeration	4
Settling	5
Impaction on Walls	5
Inertial Impaction	6
Interception	7
Diffusion	8
Droplet Nucleation	9
Thermophoresis	10
Diffusiophoresis	10
Stephan Flow	10
Summary of Theory	12
Experimentation by Others	13
Off-Gas Cleanup at Argonne National Laboratory	13
Studies in Great Britain	14
Aerosol Deposition and Containment Systems	
Experiments at Hanford	16
Studies by Other Groups at ORNL	17
Work by Other Individuals	17
Nuclear Safety Pilot Plant Experiments	17
Experimental Conditions and Results	19
Aerosol Generation and Transfer to the MCV	19
Results from Noncondensing Experiments	21
Constant-Temperature Condensing Experiments	21
Conclusions	24
References	26
Appendix A. Description of Nuclear Safety Pilot Plant	33
Appendix B. Experimental Procedure for Particle Removal by Sprays in the NSPP	43

FOREWORD

The Spray and Pool Absorption Technology Program is coordinated by Oak Ridge National Laboratory for the AEC. The program involves research on all aspects of containment spray systems proposed for use as engineered safety features in pressurized-water reactor containment buildings and investigation of certain aspects of the pool pressure-suppression containment concept as applied to boiling-water reactors.

This report summarizes work on particle removal by sprays under simulated loss-of-coolant-accident conditions. The removal of elemental iodine and organic iodides are the subject of other reports by L. F. Parsly. The material presented in these reports was previously described in a series of documents prepared to specifically report program work on spray systems. The general heading of this report series is "Design Considerations of Reactor Containment Spray Systems"; these reports, listed below, all carry the USAEC Report number ORNL-TM-2412 and a part number:

I. H. Row, L. F. Parsly, and H. E. Zittel, Design Considerations of Reactor Containment Spray Systems - Part I, USAEC Report ORNL-TM-2412, April 1969.

C. Stuart Patterson and William T. Humphries, Design Considerations of Reactor Containment Spray Systems - Part II. Removal of Iodine and Methyl Iodide from Air by Liquid Solutions, USAEC Report ORNL-TM-2412, Part II, August 1969.

J. C. Griess and A. L. Bacarella, Design Considerations of Reactor Containment Spray Systems - Part III. The Corrosion of Materials in Spray Solutions, USAEC Report ORNL-TM-2412, Part III, December 1969.

L. F. Parsly, Design Considerations of Reactor Containment Spray Systems - Part IV. Calculation of Iodine-Water Partition Coefficients, USAEC Report ORNL-TM-2412, Part IV, January 1970.

J. C. Griess, T. H. Row, and C. D. Watson, Design Considerations of Reactor Containment Spray Systems - Part V. Protective Coatings Tests, USAEC Report ORNL-TM-2412, Part V, October 1970.

L. F. Parsly, Design Considerations of Reactor Containment Spray Systems - Part VI. The Heating of Spray Drops in Air-Steam Atmospheres, USAEC Report ORNL-TM-2412, Part VI, January 1970.

L. F. Parsly, Design Considerations of Reactor Containment Spray Systems - Part VII. A Method for Calculating Iodine Removal by Sprays, USAEC Report ORNL-TM-2412, Part VII, February 1970.

H. E. Zittel, Design Considerations of Reactor Containment Spray Systems - Part VIII. Boiling Water Reactor Accident Radiolysis Studies, USAEC Report ORNL-TM-2412, Part VIII, October 1970.

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B. A. Soldano and W. T. Ward, Design Considerations of Reactor Containment Spray Systems - Part IX. The Utility of Ice Cubes as an Absorbent for Gaseous Fission Products, USAEC Report ORNL-TM-2412, Part IX (to be issued)

T. L. Hebble, Design Considerations of Reactor Containment Spray Systems - Part XII. Variability of the Dose Reduction Factor for the Removal of Elemental Iodine, USAEC Report ORNL-TM-2412, Part XII, May 1971.

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Spray and Pool Absorption Technology
Program

REMOVAL OF RADIOACTIVE PARTICLES BY SPRAYS

L. F. Parsiy

Abstract

The theory of particle removal by sprays is reviewed, and the results of a number of recent experiments by others and 15 experiments in the ORNL Nuclear Safety Pilot Plant are discussed. The experimental results show that particle removal is not significant when no steam is condensing on the spray drops but is greatly enhanced when condensation occurs. A major part of the removal by condensation is due to drop nucleation on the particles and the combined effects of diffusiophoresis and Stephan flow. Drop nucleation is a transient phenomenon and appears unpredictable, but the rate of removal due to the other phenomena can be predicted within a factor of 2 or better.

Introduction

Many nuclear power reactors are provided with containment spray systems. These are emergency systems that would be used in the highly unlikely event of a loss-of-coolant accident. In most cases, the primary function of the spray would be to remove heat and condense steam in order to reduce the pressure and temperature in the containment vessel and therefore the rate of leakage from the vessel. The sprays would also remove fission products from the containment vessel atmosphere so that they would not be available to leak to the environment.

Since it was important to show that sprays would remove fission products under accident conditions, the USAEC sponsored a spray technology program that included a solution search, studies of spray solution radiolysis, corrosion studies, single-drop absorption studies, pilot plant studies of spray performance, supporting theoretical studies, etc.¹ A major part of the program was carried out at the Nuclear Safety Pilot Plant (NSPP) at the Oak Ridge National Laboratory.

Fission products released in a loss-of-coolant accident will be in the form of gases, vapors, or solid particles. The gases and vapors include the noble gases, which are almost completely unreactive, moderately reactive compounds such as methyl iodide, and highly reactive substances such as HI and I₂. Since sprays do not have a significant effect on the

noble gases, the work at NSPP was concerned with the effect of sprays on other fission products. Three relatively independent processes were involved: (1) absorption of iodine, (2) absorption of methyl iodide, and (3) collection of airborne radioactive particles. The studies of iodine and methyl iodide absorption have been reported,²⁻⁴ and this report covers collection of radioactive particles by sprays.

Theory of Particle Capture by Spray Drops

Compared with the iodine and methyl iodide absorption processes, particle capture is complex. In the absorption cases, only one substance is considered at a time, its properties are known, and the controlling factors are readily identified. The particulate aerosol, on the other hand, consists of particles of various sizes and may be composed of more than one substance. The processes involved in the removal of particles from a containment vessel atmosphere will include at least the following: agglomeration, settling, impaction on walls, inertial impaction, interception, diffusion, droplet nucleation, thermophoresis, diffusiophoresis, and Stephan flow. The first three processes, agglomeration, settling, and impaction on walls, are those which would be effective whether sprays were operating or not in any kind of atmosphere. The next three, inertial impaction, interception, and diffusion, are ones which can occur when sprays are used even if no condensation occurs, and the final four, drop nucleation, thermophoresis, diffusiophoresis, and Stephan flow, are processes which become important when condensation occurs. We have assumed that the effects of all these processes can be added together to obtain the overall effect of the spray.

A concept which enters into the discussions of several of the phenomena is that of "efficiency." This is best defined as the probability that a particle existing in a location where it can be captured by a particular drop will be captured by that drop. For inertial impaction, it may also be defined as the ratio of the effective drop cross section for capture of a particle to the projected area of the drop. In the following discussions of the various phenomena, it will be apparent that the

efficiency will usually be affected by the particle diameter and frequently by the drop diameter as well.

The removal rate constant for the particles is the fraction of the volume swept out by the spray times the spray efficiency. The volume swept out by one drop in 1 sec is

$$v = \frac{\pi}{4} d^2 u , \quad (1)$$

where v is volume swept out, d is drop diameter, and u is drop velocity. The number of drops in the containment vessel is given by

$$N = \frac{Qt_f}{\frac{\pi}{6} d^3} , \quad (2)$$

where N is the total number of drops (assumed single size), Q is solution flow (cm^3/sec), and t_f is drop fall time (sec). Thus the total volume swept out in 1 sec is

$$v_t = \frac{\pi d^2 u}{4} \frac{Qt_f}{\frac{\pi}{6} d^3} = \frac{3Qut_f}{2d} . \quad (3)$$

If we make the simplifying assumption that the drops fall vertically,

$$ut_f = h , \quad (4)$$

where h is the fall height, and

$$v_t = \frac{3Qh}{2d} , \quad (4)$$

and the fraction F of the vessel volume contacted per second is

$$F = \frac{3Qh}{2dV} , \quad (5)$$

where V is vessel volume (cm^3). The rate constant for the process, characterized by E_i , is then given by

$$\lambda_i = E_i \frac{3Qh}{2dV} . \quad (6)$$

Agglomeration

Agglomeration acts to reduce the number but not the mass concentration of airborne particles. It is of great importance in particle removal because the larger particles produced by agglomeration settle faster and have higher inertia than their precursors. On the whole the larger particles are more readily removed than their precursors.

As a result of the random thermal motion of the gas molecules, frequent collisions of gas molecules with particles occur. Momentum is transferred from the gas molecules to the particles, and irregular motion of the particles results (Brownian movement). Particles collide, and many of the collisions result in agglomeration of the colliding particles. The process is discussed by Green and Lane,⁵ Fuchs,⁶ and Zebel.⁷ The equations range from

$$-\frac{dn}{dt} = Kn^2 , \quad (7)$$

where n is particle number concentration (cm^{-3}), t is time, and K is agglomeration constant (cm^3/sec), which was originally proposed by Smoluchowski,^{8,9} to the equation proposed by Muller:¹⁰

$$\begin{aligned} \frac{\partial n(m,t)}{\partial t} = & \int_0^{m/2} K(m_1, m-m_1) n(m_1, t) n(m-m_1, t) dm_1 \\ & - n(m, t) \int_0^\infty K(m, m_1) n(m_1, t) dm_1 , \end{aligned} \quad (8)$$

where $n(m, t) dm$ means the number of particles per cubic centimeter at time t whose masses lie between m and $m + dm$, while $K(m_1, m_2)$ gives the values of the agglomeration coefficient between particles of masses m_1 and m_2 . Equation (8) has been solved numerically^{11,12} and by similarity transformations.¹³⁻¹⁷ It is so complex that no general analytical solution exists.

Agglomeration is important when the number density of the particles is very high. The order of the agglomeration constant in Eq. (7) is 10^{-10} , so the time to halve the number of particles will be about 1 sec when $n = 10^{10}$ (Ref. 5). The principal role of agglomeration is in determining the initial aerosol size distribution (aerosol particles formed from overheated fuel are agglomerates of crystallites). It also is important in the "aging" of aerosols in containment atmospheres. In a spray-protected vessel, after the initial size distribution is established, it competes with sprays in removing particles but is usually very much slower.

The simple equation proposed by Smoluchowski, Eq. (7), is a useful first approximation. Based on empirically determined agglomeration coefficients, it gives good agreement with experimental data for a number of materials. Muller's equation has been applied with fair success to predict the concentration-time behavior of simulated LMFBR-accident-produced aerosols.

Settling

The rate of particle removal by settling in a well-stirred tank is given by⁵

$$-\frac{dn}{dt} = \frac{nv}{h}, \quad (9)$$

where n is number of particles (cm^{-3}), v is settling velocity of the particles (cm/sec), h is height of particle in tank (cm), and t is time (sec). The height factor, h , may be replaced by A_F/V for a cylindrical tank, where A_F is the floor area (cm^2) and V is the tank volume (cm^3). Thus the rate constant for settling is

$$\lambda_s = \frac{vA_F}{V}. \quad (10)$$

Impaction on Walls

Impaction on the walls should not be a significant particle-removal mechanism unless turbulence in the vessel atmosphere is great. By analogy to settling the removal rate constant is given by

$$\lambda_w = v_w \frac{A_w}{V}, \quad (11)$$

where λ_w is removal rate constant for impaction on walls (sec^{-1}), v_w is deposition velocity to walls (cm/sec), A_w is wall area (cm^2), and V is vessel volume (cm^3).

Inertial Impaction

Aerosol particles may be captured by spray drops as they fall through air. Processes of this type (specifically, capture of cloud droplets by growing drops) are important in rain formation and have been studied extensively. Mason¹⁸ discussed the theory of collisions between falling water drops and reported an analysis by Langmuir¹⁹ of the trajectories of small droplets moving past a fixed sphere of radius R . The drops were considered as points moving with the velocity of the air stream. If they moved without deviation, they collided with the sphere in proportion to its cross-sectional area. However, air streamlines were deflected around the sphere, and if the droplets had no inertia they were carried around the sphere and never collided with it. Since the drops had inertia, some collisions occurred but not as many as though the drops had not deviated from their original path. The collision efficiency is defined as the ratio of the actual collision cross section to the cross section of the sphere.

A difficulty in calculating the droplet trajectories is that no equation is known that describes the flow pattern around a sphere when both the viscous and inertial terms are taken into account. Also, when drops are nearly the same size, interaction effects between them may occur.

Langmuir¹⁹ calculated the collision efficiencies for large drops falling through a cloud of smaller ones of radius r . If the flow pattern around the large drop is assumed to be given by the potential flow solution (i.e., no effect of viscous forces on the flow pattern), while the motion of the small drop is controlled by viscous forces, the collision efficiency is a function of the parameter K :

$$K = \frac{2\rho}{9\eta} \frac{r^2}{R} (V - v), \quad (12)$$

where

- ρ = density of drops (g/cm^3),
- r = radius of small drops (cm),
- η = viscosity of the medium (poises),
- R = radius of large drops (cm),
- V = velocity of large drops (cm/sec),
- v = velocity of small drops (cm/sec).

This particle parameter (K) is identical with the Stokes number used by Fuchs.⁶

The potential flow analysis indicates that inertial impaction will not occur if K has a value less than $1/12$. Thus it follows that the efficiency of drops falling in potential flow is zero for particles smaller than

$$r_{\text{crit}} = \left[\frac{3R}{8\rho(V-v)} \right]^{1/2}. \quad (13)$$

For 1000- μ -diam drops in an air-steam atmosphere at 130°C, the critical radius is 3 μ if the particle density is one. Fission-product aerosol particles are generally smaller than 3 μ .

Langmuir also analyzed the case in which the flow pattern around the drop was considered to be governed by viscous flow.¹⁹ This consistently gave lower capture efficiencies than the potential flow model. Langmuir recommended the use of this when the large drops are falling in the Stokes law regime. Spray drops are generally too large to follow Stokes law, but drops in the lower end of the size spectrum probably follow a regime intermediate between potential and viscous flow.

Interception

Since a particle has finite size, it is possible that it will collide with or touch a drop even though its center line remains in the air streamline. Ranz and Wong²⁰ have shown that the minimum efficiency for a spherical particle lying in the path of a spherical drop is given by

$$E_o = \left(1 + \frac{D_P}{D_D} \right)^2 - \frac{1}{1 + \frac{D_P}{D_D}}, \quad (14)$$

where E_o is efficiency for interception, D_p is particle diameter, and D_D is drop diameter. For small values of D_p/D_D , Hilliard and his associates²¹ showed that this could be reduced to

$$E_o = 3 \frac{D_p}{D_D} . \quad (15)$$

It follows from this that the minimum efficiency for a 1000- μ -diam drop capturing 1- μ particles would be 0.3%.

Diffusion

It has been shown that as a result of Brownian motion, particles have a diffusion coefficient calculated by

$$D = \frac{RT}{3\pi N\eta d} , \quad (16)$$

where

D = diffusion coefficient (cm^2/sec),

R = gas constant (ergs/mole $\cdot^\circ\text{K}$),

T = absolute temperature ($^\circ\text{K}$),

N = Avogadro's number, 6.02×10^{23} ,

η = viscosity of medium (poise),

d = particle diameter (cm),

or, alternatively, if the particle size is the order of the mean free path of the gas molecules, the Cunningham correction⁵ applies and

$$D = \frac{RT \left(1 + \frac{2A\lambda}{d}\right)}{3\pi N\eta d} , \quad (17)$$

where λ is mean free path of the gas molecules (cm) and A is a correction factor.

The diffusion coefficients are small and vary inversely with particle size, so diffusion tends to be important only for very small particles. In principle the diffusivity calculated in the Ranz-Marshall²² equation can be used to calculate a deposition velocity for the particles:

$$\frac{k_c d}{D} = 2.0 + 0.6 N_{Re}^{1/2} N_{Sc}^{1/3} . \quad (18)$$

Here k_c is the mass transfer coefficient, d the drop diameter, and D the particle diffusion coefficient calculated by Eq. (16) or (17); N_{Re} is the Reynolds number $d v \rho / \eta$, where v is the drop velocity and ρ the atmosphere density; and N_{Sc} is the Schmidt number, $\eta / \rho D$.

The validity of using Eq. (18) is open to question, however. It was established for gases and vapors, for which N_{Sc} has a value not far from 1.0. For particles, because of their small diffusion coefficients, the Schmidt numbers are orders of magnitude larger.

Hilliard and his associates²¹ pointed out that the efficiency for diffusion was given by

$$E_D = \frac{4k_c}{u} . \quad (19)$$

Droplet Nucleation

It is well known that condensation of vapor to droplets usually occurs on fine particles that function as condensation nuclei. Mason¹⁸ discussed this thoroughly and covered required supersaturation, effect of soluble salts, etc. Condensation will occur when the vapor pressure of the atmosphere exceeds the vapor pressure over the drop. The drop vapor pressure increases with decreasing radius and decreases with increasing concentration of dissolved substances. For any degree of supersaturation, there will be a minimum particle size that can act as a condensation nucleus. When cold sprays are introduced into a saturated air-steam mixture, local cooling occurs that can easily be enough to cause droplet nucleation. Calculations indicate that at 130°C a reduction of 0.1°C results in supersaturation of 0.3%. The equilibrium drop diameter for pure water at 130°C and 0.3% supersaturation is 0.8 μ (per Eq. 2.3 in Ref. 18). Thus insoluble particles 0.8 μ and larger function as condensation nuclei if the atmosphere is cooled by as little as 0.1°C. More subcooling would cause nucleation on smaller particles. The droplets formed in this way can grow

to diameters of several microns and then be rapidly removed by impaction with the spray.

In many of the experiments the very marked reduction in the airborne cesium concentration observed during the first minute of spraying was believed to be due to this mechanism. However, this mechanism cannot be discussed in terms of removal efficiency, since it appears to be a transient phenomenon that is only effective when spraying starts.

Thermophoresis

If a temperature gradient is set up in an aerosol, the particles move in the direction of the lower temperature due to the difference in molecular bombardment of the particle. The theory has been discussed by Fuchs⁶ and by Waldmann and Schmitt.²³ Actually, two different theories are described. One applies when the particle radius is much less than the mean free path of the gas molecules; the other if it is much greater. Obviously, this leaves a transition region in which neither theory applies. Fuchs⁶ indicates that $0.025 < r < 1 \mu$ is the transition range; that is, it includes the particle sizes of most interest in analyzing containment spray systems. Thermophoresis accounts for very little of the particle deposition in sprays relative to other mechanisms. Therefore we will not discuss it further.

Diffusiophoresis

In a concentration gradient, aerosol particles move in the direction of the diffusion flux of the heavier gas component. The theory is discussed by Waldmann and Schmitt²³ and by Fuchs.⁶ In the case at hand, since water vapor diffuses toward the drops and air away from them, diffusiophoresis imparts a velocity away from the drop surface to the particles.

Stephan Flow

When a vapor is being removed from a gas-vapor mixture by condensation, there must be a net flow of the mixture toward the surface to satisfy hydrodynamic continuity. Suspended particles are dragged along by this flow, although the velocity may well be less than for the vapor flow. Goldsmith and May²⁴ show that the particle velocity toward the surface of a drop on

which a vapor is condensing is given by

$$V_D = - \frac{m_1^{1/2}}{y_1 m_1^{1/2} + y_2 m_2^{1/2}} \frac{D}{p_2} \frac{dp_1}{dr}, \quad (20)$$

where

V_D = particle velocity (cm/sec),

m_1 = molecular weight of condensing vapor,

m_2 = molecular weight of inert gas,

y_1 = mole fraction of condensing vapor,

y_2 = mole fraction of inert gas,

p_1 = partial pressure of condensing vapor (atm),

p_2 = partial pressure of inert gas (atm),

D = diffusivity of the vapor through the gas (cm²/sec),

r = radius (cm).

This equation gives the combined effect of Stephan flow and diffusiophoresis.

The Stephan flow velocity is given by

$$U = - \frac{D}{p_2} \frac{dp_1}{dr}, \quad (21)$$

where U is Stephan flow velocity (cm/sec).

While it would be extremely difficult to follow the process in detail, since condensation on the drop is an unsteady-state process, some useful results can be obtained in terms of average values. The rate of condensation on a drop is given by

$$\frac{dW}{dt} = \pi d^2 c_1 U, \quad (22)$$

where dW/dt is condensation rate (g/sec), c_1 is concentration of condensing vapor (g/cm³), and d is drop diameter (cm). The total condensation on the drop is then

$$W = \pi d^2 c_1 \bar{U} t_f, \quad (23)$$

where $\bar{U}$ is average velocity, t_f is drop fall time, and W is total amount condensed (g), but we can also calculate W in terms of the drop mass and temperature rise:

$$\dot{W} = \frac{\pi}{6} d^3 \frac{\rho_D c_p \Delta T}{\ell_v}, \quad (24)$$

where ρ_D is density of drop (g/cm³), ΔT is temperature rise (°C), ℓ_v is molar latent heat of vaporization (cal/g), and c_p is heat capacity (cal/g·°C). By eliminating $\dot{W}$, we find that

$$\bar{U} = \frac{d \rho_D \Delta T}{6 c_1 \ell_v t_f}. \quad (25)$$

Then from Eqs. (20) and (21)

$$V_D = \frac{m_1^{1/2}}{y_1 m_1^{1/2} + y_2 m_2^{1/2}} \frac{d \rho_D c_p \Delta T}{6 c_1 \ell_v t_f}, \quad (26)$$

and since

$$\lambda = \frac{6 V_D Q_f^+}{V d}, \quad (27)$$

it follows that

$$\lambda = \frac{m_1^{1/2}}{y_1 m_1^{1/2} + y_2 m_2^{1/2}} \frac{Q \rho_D c_p \Delta T}{c_1 v V}. \quad (28)$$

Equation (28) shows that the particle-removal rate constant due to diffusiophoresis plus Stephan flow can be calculated fairly readily if the temperature rise of the spray drops is known.

Summary of Theory

There are ten processes that may contribute to the removal of particles from an air-steam mixture by sprays. For the particle sizes and

concentrations expected to exist in a containment building atmosphere following a loss-of-coolant accident, only four of these processes should make a significant contribution to the overall particle-removal rate. These are interception, diffusiophoresis, Stephan flow, and droplet nucleation. The last of these, droplet nucleation, appears to be a transient effect. Therefore it is difficult to treat quantitatively. The other three are proportional to flow per unit volume, as is also true of absorption processes for iodine and methyl iodide. The removal rate constant for interception is inversely proportional to drop size. The rate constant for diffusiophoresis-Stephan flow is independent of drop size and depends on the amount of condensation on the drops.

Thus it appears possible to make conservative estimates of particle removal by sprays in a fairly simple way. Conservatism results from not considering some of the important mechanisms.

Experimentation by Others

A number of experiments done in Great Britain and at several installations in the U.S. on particle collection by sprays are reported in the literature. In many cases the spraying was done at room temperature, and only the impaction and interception mechanisms were important. A number of expedients were adopted to increase the impaction efficiency, including using high drop velocities and multiple contacts between the gas being cleaned and the spray; for example, by providing several sets of spray nozzles along a gas duct.

Off-Gas Cleanup at Argonne National Laboratory

In 1955, Liimatainen and Meecham²⁵ reported on experimental studies of the performance of a spray system for cleaning laboratory off-gas. Their work included two runs in which NH_4Cl aerosol was scrubbed and one for removing Na_2O aerosol. The results are given in Table 1. They reported number mean diameters of 0.5μ for the NH_4Cl aerosol and 0.037μ for the Na_2O aerosol based on determinations from electron micrographs. The aerosol concentrations were relatively low (approximately $10^3/\text{cm}^3$ for the NH_4Cl aerosol and $10^4/\text{cm}^3$ for the Na_2O aerosol). The data indicate surprisingly good removal rates for both aerosols.

Table 1. Removal of Aerosols in a Spray Tower

	Run 1	Run 2	Run 3
Length of tower, ft	15	15	10
Aerosol	NH ₄ Cl	NH ₄ Cl	Na ₂ O
Mean particle diameter, μ	0.5	0.5	0.1
Gas flow, lb/hr·ft ²	900	1250	460
Liquid flow, lb/hr·ft ³	109	109	150
Residence time, sec	4.48	3.21	5.83
Inlet concentration, particles/ft ³	79×10^6	107×10^6	511×10^6
Inlet mass concentration, ^a g/ft ³	805×10^{-7}	1135×10^{-7}	4×10^{-7}
Exit concentration, particles/ft ³	34×10^6	60×10^6	333×10^6
Efficiency of removal, %	57	44	35
$k_c \alpha$, hr ⁻¹ (b)	680	648	265

^aSpherical particles assumed.

^bMass transfer coefficient on a volume basis; units are lb-moles/hr·ft³ per lb-moles/ft³, which reduce to hr⁻¹. This is used for devices such as packed columns and spray columns in which the area cannot be determined.

Studies in Great Britain

Much work on aerosol removal has been done in various laboratories of the UKAEA, as well as elsewhere in Great Britain. Goldsmith and May²⁴ and their co-workers have carried out a number of experiments that provide information on some of the important transport mechanisms involved in particle removal by sprays, such as thermophoresis and diffusiophoresis. The reference cited contains a good discussion of their work.

Walton and Woolcock²⁶ studied the dust collection efficiency of water drops as a function of drop diameter, drop velocity, and particle diameter. This was done by suspending a drop on the end of a silicone-coated capillary in a wind tunnel and flowing air containing uniform-sized particles of methylene blue past it. A photometer was used to follow the color change of the drop. The collection efficiency was found to be a function of the parameter

$$K = \frac{20_1 \alpha^2 U}{9\eta R},$$

where

a = particle radius (cm),

ρ_1 = particle density (g/cm³),

U = drop velocity relative to particle (cm/sec),

η = viscosity of the fluid surrounding the drop and particle (i.e., the air in the wind tunnel),

R = drop radius (cm).

The results obtained were between the values predicted by Fonda and Herne for potential flow and viscous flow.²⁷ Walton and Woolcock carried out a full factorial experiment with three drop radii, three air velocities, and two dust radii, and they made two experiments at each of the 18 conditions.²⁶ They did not report an analysis of variance. They applied their single-drop results to determine the requirements for removing airborne dust from mine ventilation air and concluded that 0.5-mm (500- μ) diameter drops would be optimum (based on minimum water usage and 90% dust removal). They stated that sprays falling under gravity were not adequate, but sprays injected at high velocity by a pressure nozzle could have useful efficiency (20 to 50%).

Picknett²⁸ measured the efficiency of 30- and 40- μ -radius pure water drops for collecting smaller drops (1- to 9- μ radius) of a saline solution. The concentration of saline solution drops in the test vessel was kept low enough so that there would be a negligible probability of more than one collision occurring. The amount of salt collected was used to determine the size of the salt solution droplet. Picknett found that with a drop radius of 30 μ , 196 out of 24,083 collector drops contained salt; with an 80- μ drop, 732 out of 14,973 contained salt. He also found that for a given-size collector drop, there is a critical drop size below which effectively no coalescence occurs. The experimental collection efficiencies were roughly 10% below collision efficiencies calculated by Hocking.²⁹ Picknett was unable to decide whether this could be accounted for by experimental errors or whether it meant that about 10% of the collisions did not result in coalescence.

Pemberton has presented a theoretical analysis of the capture of nonwetttable particles by drops.³⁰ He assumed that capture can occur only

if the particle approaches the drop with enough kinetic energy normal to the drop surface to penetrate the "skin" produced by surface tension, and he made calculations to substantiate this. Then, using statistical techniques, he calculated the washout for a rainfall rate of 2.5 in./hr. His calculations indicated that nonwetable particles must be greater than 6 μ in diameter to be removed at all and that the efficiency will always be much lower for nonwetable particles than for wettable ones.

Aerosol Deposition and Containment Systems Experiments at Hanford

Much significant work has been done at Hanford by the aerosol research group under Schwendiman and by the staff of the containment systems experiment (CSE) facility. The work by Schwendiman's group on deposition from turbulent gas streams³¹⁻³⁴ and on deposition in the presence of condensing steam^{35,36} has provided valuable information on two of the deposition mechanisms. The work of the containment systems experiment (CSE) group is, however, probably of greater importance.

The CSE facility has a 25-ft-diam 30,000-ft³ pressure vessel in which eight spray tests have been carried out; runs A3, A4, A6 through A10, and A12. The first two were made at ambient temperature before the pressure vessel was insulated. The other six were made at 250 to 260°F, with the temperature being attained and maintained by adding steam to the vessel atmosphere. The results were reported in the open literature³⁷ and in project reports.^{21,38,39} An effort was made to compare observed washout coefficients with theoretical predictions based on diffusion, diffusio-phoresis, interception, and impaction being the transport mechanisms involved. The observed coefficients were significantly larger than those predicted. The experimenters consider the explanation to be either that one transport mechanism was underestimated or incorrect particle size data were used. The particle size was measured with a cascade impactor, and liquid droplets were evaporated as they passed through the instrument. A third possible explanation for the difference is that not all significant transport mechanisms were considered. Two mechanisms that may be important are droplet nucleation and eddy diffusion.

Studies by Other Groups at ORNL

Containment building sprays have been studied by several groups at ORNL. Davis⁴⁰ prepared a review of particle technology applicable to reactor safety problems, and among other subjects, he dealt with the effect of condensed moisture on particle clumping and growth. He showed that condensation of the water should be expected to accelerate particle growth and removal.

Parker, Creek, and Martin⁴¹ tested containment sprays at the end of three natural-effect runs in the containment research installation (CRI). In these runs, simulated fission-product aerosols were injected into the CRI containment vessel, and their aging behavior was observed for 20 hr. Then, sprays in the vessel were operated for several short periods of time totalling 5 min. Significant removal of iodine was observed, but there was no effect on removal of antimony, tin, or cobalt. The report does not specify important spray parameters, such as nozzle type and size, number of nozzles, and flow. The sprays were not used until most of the steam had condensed, and the combination of long aging, which would eliminate the larger particles by settling, and no condensation provided the worst possible conditions for the spray system. That is, the remaining particles were in the least favorable size range, and the important removal mechanisms associated with condensation were not available. The poor performance of the sprays is not surprising.

Work by Other Individuals

Several experimenters have studied mechanisms of particle collection by drops or other processes that may be important. Kneen and Strauss published a theoretical review and analysis of dust deposition from turbulent gas streams.⁴² Kerker and his associates studied the efficiency of 1.2- to 22- μ -diam water drops for collecting NaCl aerosol particles 0.4 to 0.8 μ in diameter.⁴³ Their results indicate 7% efficiency.

Nuclear Safety Pilot Plant Experiments

The nuclear safety pilot plant (NSPP) flowsheet for the particle-removal experiments was essentially that given in Fig. 1, with the

Fig. 1. Nuclear Safety Pilot Plant Flowsheet.

difference being that a UO_2 melting furnace replaced the iodine vaporizer. The principal component of the plant is the 1350-ft³ model containment vessel (MCV). Other major components are the 100-gal solution storage tank, the solution pump, and the UO_2 melting furnace. A detailed description of the facility is given in Appendix A.

Appendix B describes the experimental procedure in detail. Briefly, a $\text{UO}_2\text{-Cs}_2\text{CO}_3$ pellet was melted with a plasma torch and the aerosol generated was swept into the MCV. At the time the aerosol was released the MCV usually contained a simulated loss-of-coolant-accident atmosphere (a mixture of air and steam). After a short delay to homogenize the MCV atmosphere and to obtain starting samples, spraying was started. Samples were taken at intervals during the spraying period (normally 1 hr), and final samples were obtained after spraying was stopped. The MCV atmosphere was then purged of the remaining aerosol, and the sampling devices located in and on the vessel were removed. Next a decontamination procedure was followed to remove essentially all the cesium and uranium left in the MCV and the furnace. All data and samples needed to maintain a complete material balance were collected.

Experimental Conditions and Results

The experimental conditions and results of the 15 particle-removal runs are listed in Table 2. In 13 of the runs steam was condensed, and in the other two, condensation was prevented — once by running at ambient temperature and the other time by spraying hot water.

In ten of the condensing runs, no steam was added after the specified initial temperature and pressure were attained. In this group of runs, the containment vessel temperature decreased from 130°C at the start of the spraying period to approximately 50°C at the end. The circulating solution was cooled as much as possible by a coil in the solution storage tank and a jacket on the line between the pump and the MCV.

In the remaining three runs, steam was added continuously to maintain the initial temperature. Two runs were made at 120°C and one at 100°C. We chose 120°C rather than 130°C as the upper temperature for these runs to allow some leeway for temperature variation around the set point. Our experience showed this was unnecessary; the temperature was controlled within 1°C in all three runs.

Table 2 shows that we were able to obtain more complete and consistent results for cesium than for uranium. We believe the principal reason was analytical sensitivity. The uranium samples frequently contained only a few micrograms; that is, just about the minimum quantity of uranium that could be determined at all. While the mass concentration of cesium was frequently lower than that of uranium, the samples generally contained enough tracer to give good counting statistics. For this reason, the discussion below is based on the results obtained for cesium.

Aerosol Generation and Transfer to the MCV

Good release of material from the fuel was obtained in all cases. The cesium release ranged from 50 to 90% of the inventory in the fuel, and the uranium release was from 10 to 15%. However, only about 10% of the released material was transferred to the MCV, and the remainder was deposited in the furnace. Further loss of aerosol invariably occurred in the MCV, since the concentrations found in the first gas samples taken represented

Table 2. Conditions and Results of NSPP Particle-Removal Experiments

	Run 55	Run 59	Run 60	Run 61	Run 62	Run 63	Run 64	Run 67	Run 68	Run 70	Run 71	Run 83	Run 84	Run 85	Run 86
Number of spray nozzles	3	3	3	3	3	3	3	6	12	12	1	1	1	1	1
Type of nozzles	703 ^a	703	703	703	703	703	703	A-8 ^b	A-8	A-8	1713 ^c	1713	1713	1713	1713
Solution flow, gpm	11	11	11	11	11	11	11	7.1	14.2	14.2	14	14	14	14	14
Initial vessel pressure, psig	45	3	45	22	45	45	7	45	42	45	47	38	30	30	17
Initial vessel temperature, °C	130	27	130	100	130	130	70	130	130	130	132	129	120	120	102
Initial solution temperature, °C	26	22	25	15	23	17	18	21	22	24	24	23	23	23	24
Spraying time, min	50	50	60	60	60	5	60	60	60	60	60	60	60	60	60
Steam added while spraying	No	No	No	No	No	No	No	No	No	No	No	No	Yes	Yes	Yes
Cesium released and found, mCi															
Released from sample	188.6	126.8	96.0	212.6	172.5	132.7	194.7	95.0	107.9	165.8	52.5	71.1 ^d	112.7	107.7	116.11
Transferred to MCV	16.728	11.010	11.27	35.31	24.71	12.99	30.32	7.13	13.32	13.581	10.361	6.737	14.12	12.76	7.20
Airborne in initial gas sample	7.44	11.78	32.17	18.4	5.19	5.39	21.43	2.42	3.35	11.0	2.29	4.59	5.83	1.65	4.20
In spray water	15.001	5.622	10.042	30.75	18.74	11.11	24.75	6.22	10.17	12.867	4.012	6.19	11.13	10.39	6.05
In MCV decontamination solution	1.572	1.207	1.117	3.83	2.87	1.663	5.03	0.872	3.14	0.652	6.317	0.534	2.88	2.39	1.15
In purge	0.128	4.18	0.109	0.734	3.10	0.216	0.541	0.041	0.011 ^d	0.17	0.032	0.0659 ^d	0.1475 ^d	0.096 ^d	0.0373 ^d
Overall cesium decontamination factor	131	2.6	103	48	8.0	60	56	174	121.0	80	24	102	95	133	193
Decontamination factor for spray	53	2.8		25	1.7	24	40	59	305	65	72	70	40	17	113
Uranium released and found, mg															
Released from sample	5166	6252	875.2	6855	3353	4559	6629	1799	851.7	3025	544.0	337.6	1602	901.8	197.6
Transferred to MCV	310	124.09	107.14	452.0	322.7	393.2	1178	92.15	125.84	271.46	50.49	50.41	154.3	82.68	30.04
Airborne in initial gas sample	109.5	83.9	3.48	1187	201	1072	3720	5.63	49.0	158	6.55			9.54	
In spray water	16.25	29.11	43.13	228.0	18.9	75.36	445.7	5.59	25.57	13.72	12.53	0	79.16	38.31	7.45
In MCV decontamination solution	329.76	88.58	62.79	201.0	282.8	309.3	716.5	86.45	100.33	255.63	37.96	21.29	75.12	44.20	22.59
In purge	19.2	6.40	0.218	23.0	21.0	8.54	15.8	0.109	0.115 ^d	2.11	0.672	29.12	12.8	0.17	
Overall uranium decontamination factor	18	19	49.1	19.7	15.4	46	75	845	1090	129	75	1.7	12.1	486	
Decontamination factor for spray	5.7	13.1	16.0		9.6			52	426	75	9.7			56	

^aSpraying Systems Type 703 is cluster of seven size 03 nozzles manufactured by Spraying Systems Company, Bellwood, Illinois.

^bSpraying Systems Type A-8 is hollow-cone nozzle manufactured by Spraying Systems Company, Bellwood, Illinois.

^cSpraco Type 1713 nozzle manufactured by Spray Engineering Company, Burlington, Massachusetts.

^dCalculated from gas samples taken after spraying.

only 10 to 40% of the aerosol transferred, as determined by the material balance.

Results from Noncondensing Experiments

In runs 59 and 62, condensation was essentially eliminated. In run 59 the containment atmosphere was left at room temperature, while in run 62 the atmosphere was heated to loss-of-coolant-accident conditions, but the spray solution was also heated. In both runs the decontamination factors measured for cesium were low. In run 59, it was 2.8, and based on the relationship

$$DF = e^{\lambda t}, \quad (29)$$

the removal rate constant was $3.43 \times 10^{-4}/\text{sec}$. The mean particle size of the aerosol was 0.7μ before spraying and 0.3μ after. The removal rate constant estimated by using Eqs. (15) and (6) was $1.5 \times 10^{-4}/\text{sec}$, in fairly good agreement with the experimental results.

In run 62, the decontamination factor attributable to the spray was 1.7, indicating a removal rate constant of $1.5 \times 10^{-4}/\text{sec}$. The particle size in this run was 1.7μ before spraying and 0.47μ after. The calculated removal rate constant was $3.4 \times 10^{-4}/\text{sec}$.

Constant-Temperature Condensing Experiments

The final three experiments were run with steam being added continuously while spraying. This type of experiment is inherently simpler than experiments in which steam is not added because the containment atmosphere temperature is constant during the experiment.

Figure 2 shows how the airborne cesium concentration varied with time. Only 58% of the cesium delivered to the containment vessel was found airborne by gas sampling before spraying started. As soon as the sprays were turned on, the cesium concentration dropped by a factor of over 10. Based on observations of the gamma-ray intensity at the MCV wall, we believe this occurred in about 1 min. During the remainder of the 1-hr spraying time the concentration decreased by an additional factor of about 11. A further

Fig. 2. Normalized ^{137}Cs Concentration in MCV Atmosphere Versus Spraying Time in NSPP Run 86.

decrease by a factor of about 10 occurred during the waiting period between the end of spraying and the start of purging.

We think the rapid concentration decrease when spraying starts is due to growth of drops on particles. The drops grow rapidly to sizes where they are rapidly removed by settling, interception, and impaction.

During the remainder of the spraying time, removal continues at a rate of approximately 2.5/hr. We believe this represents the combined effect of interception and diffusiophoresis-Stephan flow. Additional removal between the end of spraying and purging occurred at an average rate of about 0.125/hr. This is partly due to condensation as the vessel cools; however, we believe that more removal occurred than can be accounted for by condensation.

The development in the theory section of this report indicates that the rate during the second particle-removal period should be predictable

from the data on the experimental conditions. For the prediction we assumed that only interception and diffusiophoresis-Stephan flow made significant contributions to the particle-removal rate. Figure 3 shows the removal while spraying for runs 84 and 85, run 86, and the earlier runs in which no steam was added while spraying so that the vessel cooled continuously. From these data we calculated a removal rate constant of $7.86 \times 10^{-4}/\text{sec}$ for runs 84 and 85 and a constant of $7.00 \times 10^{-4}/\text{sec}$ for run 86. The constant due to interception was calculated to be $1.2 \times 10^{-5}/\text{sec}$. Hence the constant due to diffusiophoresis-Stephan flow was $7.74 \times 10^{-4}/\text{sec}$ for runs 84 and 85 and $6.88 \times 10^{-4}/\text{sec}$ for run 86. Based on the spray solution temperature rise and flow the expected removal rate constants were 9.2×10^{-4} and $1.12 \times 10^{-3}/\text{sec}$, respectively. Both are within a factor of 2 of the observed constants. The other removal phenomena do not appear to be predictable and must be viewed merely as "plus" factors.

In the decreasing temperature runs, the average rate while spraying (excluding the initial drop attributed to droplet nucleation) yields a constant of $5.13 \times 10^{-4}/\text{sec}$ or about $5.01 \times 10^{-4}/\text{sec}$ after the interception contribution is subtracted. The total condensation would be 37% of that

Fig. 3. Cesium Removal While Spraying in Condensing Runs.

in runs 84 and 85, and thus the "expected" constant would be 37% of that for runs 84 and 85. The observed constant was considerably more than that. This again suggests that significant particle removal must be associated with cooling of the containment atmosphere.

Conclusions

The experimental results indicate that not much particle removal occurs unless steam is condensing on the spray drops, in which case two phenomena appear to be important — drop nucleation, which apparently occurs when spraying starts, and diffusiophoresis-Stephan flow, which continues as long as there is condensation on the drops. We do not believe that the reduction in particle concentration due to drop nucleation is predictable enough to permit its being taken into account in an accident analysis. On the other hand, the discussion above indicates that the rate of particle removal by the combined action of diffusiophoresis and Stephan flow is predictable within a factor of 2. Therefore it should be possible to claim that one-half the calculated removal rate constant is suitably conservative.

Equation (28) for calculating the removal rate constant due to diffusiophoresis plus Stephan flow shows that the constant depends on the spray flow per unit volume and on the temperature rise of the drops. The flow per unit volume is an adverse factor for scaling, since in typical containment buildings it is 8 to 15% of that used in the NSPP experiments. On the other hand, the temperature rise of the spray drops in containment buildings should be two or three times that in the NSPP. Thus the removal rate constant due to the condensation-related effects should be 20 to 40% that in the NSPP experiments. While this seems small, it is enough to reduce the 2-hr integrated dose contribution from particles by a factor of about 1.7, which should be significant.

The results presented here show that the concentration of airborne particles will be reduced significantly by sprays under loss-of-coolant accident conditions. A conservative estimate of the removal rate can be made from data on spray flow and temperature rise, and we recommend using one-half the calculated rate constant.

Additional experiments in which the condensation rate was varied over a much greater range would be desirable. The rate in the NSPP experiments was limited by the ability of existing equipment to cool the recirculated spray solution. It would have been necessary to add a heat exchanger to the NSPP system and to provide more solution storage volume to produce a truly variable condensation rate in the NSPP. A properly planned program should be able to obtain data which would considerably increase confidence in our ability to predict particle removal by sprays.

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APPENDICES

Appendix A

DESCRIPTION OF NUCLEAR SAFETY PILOT PLANT

The facility for the particle-removal experiments consisted of the model containment vessel (MCV), the sample-melting furnace, solution preparation tank, solution storage tank, solution circulating pump, solution weigh tank, and the sampling equipment. The flowsheet is shown in Fig. 1 in the main body of this report.

Model Containment Vessel

The MCV is a 10-ft-diam, 15-ft straight side, vertical, cylindrical, stainless steel pressure vessel with ASME flanged and dished heads. It was designed and built in accordance with Section VIII of the ASME Boiler and Pressure Vessel Code for 60 psig at 300°F internal pressure and full vacuum. Figure A.1 shows the vessel.

Furnace

The furnace used for the particle-removal experiments is illustrated in Fig. A.2. The body was stainless steel, and the crucible for the sample was graphite. To assure cooling the furnace was submerged in a tank of water with the water surface at least 1/2 in. above the top flange. The spacing between the torch face and the sample could be adjusted by shimming under the mounting lugs on the torch.

The torch was an ORNL modification of an AVCO plasma gun, Model PG-100. The modified design was easy to assemble without internal leaks; it had O-rings on a cool section of the torch body to make the seal between the torch and the furnace lid. Current for the torch was supplied by one AVCO PS 350-1 master unit and two PS-352-1 slave units connected in parallel. These provide up to 1050 A at 40 V. The units were essentially arc-welding power supply units comprising a transformer, a self-saturating reactor, and a selenium rectifier. They were connected to the torch by water-cooled cables, and the water that cooled the cables was also used in the torch to cool the electrodes.

Fig. A.1. Model Containment Vessel of Nuclear Safety Pilot Plant.

Fig. A.2. Furnace for Melting UO_2 for Particle-Removal Experiments in NSPP.

Helium was supplied to the torch to form the plasma. It was normally injected tangentially just downstream of the torch face, since the vortex so formed seemed to stabilize the plasma jet.

The furnace had two view ports. One was used for observing melting, and the other was for a pyrometer.

Solution Preparation Tank

The solution preparation tank was an open-top 55-gal stainless steel drum mounted on the working floor above the hot cell where the pilot plant equipment is installed. It had a portable mixer. All solutions used were prepared in this tank and drained by gravity to the solution storage tank in the cell.

Solution Storage Tank

The solution storage tank was a horizontal cylindrical stainless steel pressure vessel of 100 gal capacity. It contained a coil that could be supplied either with steam to heat the tank contents or with cooling water. It served as a working storage volume for either spray or decontamination solution.

Solution Circulating Pump

The solution circulating pump was an Allis-Chalmers type DGH, Model C-11BS, horizontal, centrifugal, canned-motor pump, and all parts in contact with the fluid were type 316 stainless steel. The pump was rated at 50 gpm at 120 ft, and it had a 5-hp 3500-rpm motor.

Solution Weigh Tank

The solution weigh tank was a vertical, cone-bottom, cylindrical, stainless steel tank suspended from a weigh cell. It was equipped with an air sparger to mix its contents. The tank and contents could be weighed to within 0.55 kg by the weigh cell. Samples could be withdrawn from near the apex of the cone bottom.

Sampling Equipment

Equipment was provided for collecting samples of the MCV atmosphere, spray solutions, etc., to enable behavior in the system to be followed. The samplers included May-pack clusters and low-pressure cascade impactors

for sampling the containment atmosphere. spray solution samplers, wall run-off samplers, and sump samplers. All sampling was remotely controlled from the NSPP control room.

May-Pack Samplers

The May pack is a device in which a series of different collection media are installed. It was originally devised by F. G. May of UKAEA to collect and differentiate iodine forms. A typical May pack is illustrated in Fig. A.3. It contains in sequence absolute filters to collect particles, silver-plated screens to collect I_2 , and charcoal-impregnated fiber-glass plus charcoal beds to collect other iodine forms. Since for the particle-removal experiments only the particles were of concern, the pack was reduced to two absolute filters. The May packs were installed in devices called May-pack clusters (Fig. A-4). Each cluster contained 12 May packs and a modified Gelman sequential sampling valve to permit these packs to be sequentially sampled. To keep iodine from getting to the collection media except while a sample was being collected, a spring-loaded ball check valve was installed at the inlet to each May pack. For these particle experiments, however, we decided that we could hang the May packs vertically with the inlets facing down and omit the check valves, which we had found to be very good particle collectors.

We considered it important to get a good measurement of the sample size, and we did that in the following manner. The sampling line from the May-pack cluster ran down and out a penetration in the lower part of the MCV through a throttling valve to a condenser in a chilled-water bath. Then it ran through a glass-wool plug for mist removal and was metered by a calibrated capillary flowmeter. The inlet absolute pressure and differential pressure of the capillary were sensed by transmitters and indicated in the control room. Air loading to the throttling valve was adjusted manually at the sampling panel.

Since the flow metering system measured the flow of the air contained in the sample and the density of air in the MCV was practically constant as long as the atmosphere was well mixed, the volume of air measured by the metering system was the volume of MCV atmosphere in the sample.

ORNL DWG 67-11058

Fig. A.3. May-Pack Assembly Used for Particle-Removal Experiments in NSPP.

Fig. A.4. May-Pack Cluster.

The condensate and the glass wool plug were saved, and their activities were counted. A large amount of activity would have indicated that the sampling channel was defective and would have been a basis for discarding the data from the May-pack cluster in the channel.

Low-Pressure Cascade Impactors

Since particle size is one of the most important properties of an aerosol, data on size were necessary for evaluating the experimental results. The type of aerosol with which we were dealing was formed by condensation of vapors into crystallites followed by agglomeration of the crystallites to form the aerosol particles. The size and shape of the crystallites probably depended on the vapor concentration and temperature gradient where condensation took place. Typically, they were of the order of 100 Å in diameter. These agglomerated into chains and clumps that contained up to several hundred crystallites. It was the properties of the agglomerates that were important in determining aerosol behavior.

Cascade impactors are useful devices for measuring the aerodynamic size of aerosols. However, impactors operating at atmospheric pressure are limited to collecting particles 0.5 μ and larger in diameter. To overcome this limitation, the impactor can be run at reduced pressure. Parker and Buchholz* modified a commercially available impactor for low-pressure operation (Mark I) and subsequently built an improved version (Mark II). The Mark I unit and one Mark II unit were available at the NSPP and were used to analyze one sample of the aerosol before spraying started and one after spraying was stopped.

Off-Gas Sampling

In order to complete the material balance and to determine the activity at the end of a run, a sample was collected of the air vented and purged after each run. The air was released at a fixed rate (30 cfm),

*G. W. Parker and H. Buchholz, Size Classification of Submicron Particles by a Low-Pressure Cascade Impactor, USAEC Report ORNL-4226, Oak Ridge National Laboratory, 1968.

and a metered sample of the air being released was drawn through a single May pack. Makeup air was added through the jet of the model containment vessel draft tube, and thus it functioned to induce rapid circulation of the vessel atmosphere and became intimately mixed with the atmosphere.

Solution Sampling

Facilities were provided for collecting three different types of solution sample. One was the solution that had fallen through the MCV atmosphere. The second was liquid from the pool at the bottom of the MCV. The hydrodynamic characteristics of the NSPP were such that about one-half the solution remained in the pool at the bottom of the MCV when solution was being recirculated. The third was solution (plus condensate) that reached the walls and ran down them. These samples are referred to as the "spray sample," the "sump sample," and the "wall sample."

Spray Sample

The spray sample was collected in a trough made by slitting a 3-in. pipe lengthwise. The trough extended along a radius from the center of the MCV practically to the wall. It was drained to a manifold and collected in one of six bombs. There was provision for flushing the header with solution so that the sample in the bomb was a true sample of the solution at the time it was collected.

Sump Sample

To get a sample of the solution in the MCV sump, a sidestream was taken off the vessel draw-in line and returned to the sump. A small Chempump circulated the solution and helped mix the sump contents. A section of the recirculation line had five parallel paths, four of which could be isolated by closing valves at both ends. The fifth provided a continuously open flow channel. When a sample was to be taken the valves on one of the four sections were closed. After a run, the samples were drained into bottles.

Wall Sample

A trough 4 in. wide was attached to the wall to collect a portion of the solution running down. This was connected to a six-bomb manifold similar to that for spray sampling.

Appendix B

EXPERIMENTAL PROCEDURE FOR PARTICLE
REMOVAL BY SPRAYS IN THE NSPPPreparation

The preparations for a run comprised sample preparation, assembly and installation of sampling devices, installation of spray nozzles, leak tests, sample loading, and establishing initial conditions. All the samples were prepared in a single run by the ORNL Isotopes Division. Cesium-137 was the only radioisotope used, so there was no need to prepare samples immediately before use. A sample is shown in Fig. B.1 after melting in the furnace. The central part is a right cylinder cold-pressed from a mixture of UO_2 and Cs_2CO_3 proportioned to give the cesium-to-uranium ratio present in reactor fuel after 20,000-MWd/MT exposure. Enough ^{137}Cs was added to give 200 mCi in each sample, since experience had shown that this was a convenient sample activity level for the NSPP.

The $\text{UO}_2\text{-Cs}_2\text{CO}_3$ pellet was enclosed in Zircaloy-2 cladding made of tubing with press-fit end plugs. This in turn was enclosed in a stainless

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Fig. B.1. Sample for Particle-Removal Experiment Shown After Melting in Furnace.

steel outer cladding, which was welded to assure against spills while handling.

The sampling installation consisted of two May-pack-cluster gas-sampling devices installed inside the containment vessel, a device to collect six samples of spray solution from near the bottom of the containment vessel, one to collect six samples of wall runoff, one to collect four samples of the liquid in the bottom of the containment vessel, two lower pressure cascade impactors, and a single May pack for collecting a sample of the air purged after the run.

The May-pack samplers, which may be loaded with a variety of collection media, as mentioned in Appendix A, contained only filters for the particle-removal experiments. For iodine experiments, inlet check valves are normally installed, but for the particle-removal experiments, the check valves were left out and the samplers were hung so that the inlets faced down and no particles would get on the filters unless a sample was actually being taken.

The May-pack samples were taken by drawing approximately 1 liter/min from the MCV through a sample tube by means of a sampling vacuum pump located outside the MCV. En route to the vacuum pump the sample passed through a throttling valve, a condenser located in a chilled-water bath, and a metering capillary. Either 5- or 10-liter samples were taken. The capillaries were calibrated over a wide range of inlet and differential pressures, which were measured as the sample was being taken and read out on the control panel. Since only the air content of the gas sample was measured, the volume of the sample as metered was the volume of MCV atmosphere represented by the sample. (The MCV contained a fixed amount of air, and no matter what was done to the temperature and water vapor content, so long as the atmosphere was well mixed, the density of air in the vessel remained essentially constant. The total volume of the gas samples was less than 0.5% of the MCV volume and hence was not significant.)

After all the sampling devices were cleaned, assembled, installed, and checked functionally, the MCV was closed and pressurized to 45 psig for a leak test. The pressure and temperature was monitored for at least 16 hr, and the volume of contained air at the beginning and end of the

test period were calculated. Leakage of up to 1% per 24 hr was considered acceptable. If there was more leakage, the major leaks were found and repaired, and the test was repeated.

Once the leakage was acceptable, the vessel was depressurized and the sample loaded into the furnace. This was done by removing the plasma torch and placing the sample in the graphite sample holder with tongs. The torch had a double O-ring seal that could be monitored and was easy to make leak-tight. After the sample was loaded, a final pressure leakdown test was made. A run plan was also prepared that indicated all operations to be done and included a detailed sampling schedule.

The required volume of water was then loaded into the solution storage tank. (After reviewing the matter with our consultants, it was determined that there was no reason to use spray solution.) Finally, on the day the run was to be made, the MCV was heated with open steam. To keep steam out of the furnace, a slight helium purge was used.

Aerosol Generation

Once the system was at temperature, the condensate formed while heating was drained, and the aerosol was generated by melting the sample with the plasma torch and sweeping the vapors into the MCV with the gases associated with torch operation. A 10-min melting period was used in most runs. The draft tube used to mix the contents of the MCV was operated continuously in some of the runs. Eventually, we concluded that the draft tube caused too much aerosol loss and used it only during the last 2 min of torch operation.

Spraying

After making the aerosol, we took starting samples. These included a May-pack sample for concentration and a cascade impactor sample for particle size. In all but two runs, spraying was started at this time. In those two, a 1-hr natural-effects period intervened before spraying started, and a second May-pack sample was taken at the end of the natural-effects period. In all runs the sprays were operated for 1 hr, which was

thought to be long enough to give adequate data. Two gas samples were taken while spraying and a final gas sample was taken starting 5 min after the sprays were turned off. The second cascade impactor sample was taken at the same time.

The water initially was cooled as much as possible and maximum cooling was continued during the runs. The cooling was provided by a helical coil in the solution storage tank, which was normally only 50% submerged when spraying, and a jacket on the 1 1/2-in. pump discharge line. These could cool the water about 20°C.

In most runs the temperature was allowed to decrease during the spraying period; however, in the last three runs steam was added to maintain a constant temperature.

Samples of the solution in the bottom of the MCV, the solution running down the vessel wall, and that which had fallen through the vessel atmosphere were collected at intervals while spraying.

Cleanup

At the end of the spraying period the solution was drained from the vessel and the vessel was allowed to stand overnight. The solution was then transferred from the solution storage tank to a weigh tank, where each batch was weighed and sampled. The following morning any accumulated runoff was drained. Then the MCV was purged for 4 hr at 30 cfm with dry air. The flow leaving was measured with an orifice meter, and a continuous sample was taken and accumulated on a May pack.

After purging was completed, the top flange was removed from the MCV, and the May-pack clusters were remotely disconnected and lifted out. The top flange was replaced, and then the liquid samplers were removed. All samplers were transferred to a sample processing area, where the samples were separated, packaged, and labeled for transfer to the radiochemical analytical laboratory. All samples were counted for ^{137}Cs . Those which contained significant cesium were then sent to be analyzed chemically for uranium. On the first particle run we also had analyses made for tin, which has been reported to be evolved from Zircaloy. Not enough was found to justify analyzing for it.

The furnace and MCV were then decontaminated by the following procedure. First the MCV was washed with uranium-complexing solution (10.8 lb oxalic acid, 5.6 lb ammonium citrate, 36 lb 30% hydrogen peroxide, 36 gal water, and NH_4OH as required to adjust the pH to 2.5) at 70°C. Two decontamination headers were used to provide overlapping coverage by full-cone nozzles to wet the entire top head and a series of fan spray nozzles to wet the vertical wall at the top tangent. This washing was followed by three hot water rinses. Then the plasma torch was removed from the furnace and a spraying fixture installed in its place. The furnace was rinsed thoroughly with hot water. Then the spraying fixture was removed temporarily while the sample residue was taken out. After removal of the residue, the furnace was decontaminated with the uranium-complexing solution and rinsed three times. All furnace decontamination solutions were allowed to overflow into the containment vessel through the connecting line. A final water rinse of the MCV was then made to remove any residue left by furnace decontamination. Each batch of solution or rinse water was transferred to the weigh tank, weighed, and sampled after use. All these samples were analyzed for both cesium and uranium.

The furnace decontaminating sequence was designed so that the sample residue could be removed before the complexing solution was used in the furnace. The water rinse carried out before retrieving the sample reduced the loose contamination associated with the residue.