

PHYSICAL AND CHEMICAL FACTORS INFLUENCING FISSION
PRODUCT RETENTION IN REACTOR PRIMARY SYSTEMS

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ABSTRACT

The transport of fission products in vapor and particulate form through a reactor coolant system under accident conditions is accompanied by deposition of the fission products onto system surfaces. Deposition occurs by a number of mechanisms with the predominant mechanisms being vapor condensation and reaction with surfaces, and particle agglomeration and sedimentation. Deposition of more than 90 percent of fission products in the forms CsI, CsOH, and Te have been predicted for some accident conditions.

Several computer codes have been written to perform calculations of transport and deposition and include most physical mechanisms believed to be of importance. However, there are several issues of importance to be resolved. These include processes such as chemical reactions, decay heating of fission product deposits, resuspension of deposits, and natural circulation within the primary system. It is current thinking that decay heating and chemical reactions may be important to retention at long accident times and natural circulation may significantly affect thermal hydraulic conditions. Consideration of these issues are expected to lead to revisions of the transport codes.

Under reactor accident conditions, the core heats to a point where vaporization of various materials occurs. As radionuclides, control rods, and structural materials are vaporized in the core region, they move from the vaporizing surface through a concentration and temperature gradient or "boundary layer" out into the bulk vapor. Much of the initial particle formation occurs by nucleation in this region near the vaporizing surface. The vapors and particles are then moved along through the core region with the gas flow. As this gas flow moves to cooler regions in the core or moves into downstream portions of the primary system along the leak flow path, condensation and sorption of vapors occur on particle and system surfaces, the particles grow by agglomeration and deposit on surfaces, and if the system surfaces are heated either by decay heating or by heat transfer from the gas, the deposited materials may be evaporated from the surfaces. There are other effects such as chemical interactions with the surface which may also influence the transport and deposition process. The overall behavior of the radionuclide transport can be typified as being governed by the physical and chemical processes affecting interactions among particles, vapors, and surfaces.

Fission product deposition in reactor coolant systems under reactor accident conditions is generally expected to be a significant factor affecting the radionuclide source term to the environment. This is in contrast to previous assumptions, such as in the Reactor Safety Study, where no credit was taken for attenuation in the primary system because of expected low deposition rates as well as the possibility for revaporization by decay heating. Recent calculations⁽¹⁾ have estimated that more than 90 percent of the cesium, iodine (as CsI and CsOH), and tellurium inventories may be deposited in the primary system for certain accident sequences. These amounts of deposited materials significantly reduce the amounts available for transport in the containment, and if later revaporization occurs, may lead to conditions where initial primary system deposition is of major concern as a source at later times. The important conclusion is that primary system deposition may be of major importance in either reducing the release to the containment or in changing the release time relative to containment failure time.

Considerable effort has been directed toward developing the complex codes used to predict fission product retention in reactor coolant systems. Nevertheless, there remain issues that must be resolved and code validations to be completed. It is the purpose of this paper to review the general approaches taken for analyzing primary system retention and to identify outstanding issues requiring resolution.

MODELS FOR PRIMARY SYSTEM DEPOSITION

There are basically three computer codes available for predicting the transport and retention of fission products in the reactor coolant system. These are the TRAP-MELT 2, RETAIN, and RAFT codes. Each of these treats fission products in both vapor and aerosol form. Other codes are available for predicting aerosol transport, but since vapor transport is believed to be important, the above three codes will be emphasized. Additional codes are under development (MELCOR and MELPROG) which will be employing more detailed treatments of the entire reactor system under accident conditions. A revision of the

TRAP-MELT 2 code is also in progress which will include predictions of thermal hydraulic conditions in conjunction with fission product transport.

Because it has served as a basis for developing other codes and has been used extensively in accident analyses, the general approach for code development will be discussed in terms of the TRAP-MELT 2 code and then comparisons made with other codes.

TRAP-MELT 2 Code

The TRAP-MELT 2 model is designed to treat radionuclide transport in an arbitrary flow system whose thermal hydraulic conditions are provided as input as functions of time.^(2,3) In addition, TRAP-MELT 2 requires the definition of source terms for each radionuclide in terms of mass release rates in the core region. Once the flow system is defined, it is subdivided into a series of control volumes that can, in principle, be arbitrary in number and flow connections and that are chosen on the basis of characteristic geometry, thermal hydraulic conditions, and suspected significant radionuclide behavior such as change of phase, agglomeration, or deposition. Radionuclides in each control volume are assigned, with uniform distribution, to one of two carriers: the wall surfaces and the gas phase. Each radionuclide is allowed to reside on these carriers in either particulate (liquid or solid) or vapor form so that by combining carrier with form in the concept of "state", the condition of a radionuclide in a given control volume is completely determined by its state. TRAP-MELT 2 thus considers five states:

- Radionuclide vapor carried by gas
- Radionuclide particle carried by gas
- Radionuclide vapor carried on wall surface
- Radionuclide particle carried on wall surface
- Radionuclide vapor chemisorbed on wall surface.

This list of states is not exhaustive (for instance, in two-phase flow, the carrier water must be considered) and the logic of the code has been chosen to accept an arbitrary number of states readily.

Radionuclide transport can occur among the five states of an individual control volume or between certain states of different control volumes are connected by fluid flow. The former types of transport are modeled or correlated in the code itself. The latter are assumed to occur in phase with the fluid flow and are imposed on the system. Sources of radionuclides to the system may occur in any volume and any state, and they may be input to the code as mass rate functions of time.

At present, the intravolume transport mechanisms contained in TRAP-MELT are:

- Competitive condensation on, or evaporation from, wall surfaces and particles of cesium iodide, cesium hydroxide, and tellurium

- Irreversible sorption of molecular iodine, cesium hydroxide, and tellurium on stainless steel surfaces
- Particle deposition on surfaces due to
 - Settling
 - Diffusion from laminar and turbulent flow
 - Inertial impaction from turbulent flow
 - Thermophoresis.

Particle transport (and evaporation or condensation from or on particles) depends on particle size. TRAP-MELT 2 takes this into account by considering a discretized particle size distribution that is subject to change, in each volume, by the deposition processes themselves, by possible particle sources, by flow of particles from other volumes, by flow of particles out of the volume in question, and by agglomeration. The last can be due to many mechanisms. TRAP-MELT 2 considers the following agglomeration mechanisms:

- Brownian
- Gravitational
- Turbulent (shear and inertial).

Considerations of stiffness and linearity split the system of first order differential equations resulting from the above-listed transport mechanisms into three classes. Most of the deposition mechanisms (transfer from gas to wall surface) are taken as first order in the concentration of radionuclide species on the carrier (gas, particle, or wall) from which the transfer occurs. They constitute the first class, whose transport scheme can be written in the form:

$$\frac{dC}{dt} = S + MC, \quad (1)$$

where C is the concentration vector of the species in question for each state and volume, S is the source rate vector for each state and volume, and M is the transport matrix between all states and volumes. Because the deposition terms are taken as first order, M is independent of C and depends, with S , on time only. It is thus possible to solve Equation (1) as a set of first order differential equations with constant coefficients by standard techniques. This is done in TRAP-MELT 2 for the class of linear mechanisms. Condensation and evaporation, which have a much shorter time constant than the linear processes, constitute the second class and are treated outside this framework but parallel to it, as is particle agglomeration, which constitutes the third class of mechanisms in the TRAP-MELT 2 code.

The approach to this parallel treatment is as follows: Equation (1) is taken as the master time-translation operation of the radionuclide system. Time steps are adjusted so that S and M change little over a time step and so that the time step does not exceed one-third of the smallest flow residence time for any control volume. The latter assures that the system does not translate excessively between couplings to the other two classes of mechanisms. In addition, the characteristic coagulation time for the aerosol in each volume is evaluated and compared to the master time step. If the former is short compared to the latter, the master time step is appropriately reduced.

At the beginning of each time step, phase transitions of radionuclides are modeled by examining each control volume in turn and solving the molecular mass

transport equations for vapor transport among the gas phase, particles, and wall surfaces. Because of the low heats of vaporization of the radionuclides in question, this transport is assumed to be isothermal. Transfer to the walls assumes the Dittus-Boelter correlation for pipe flow and transfer to the particles occurs by diffusion based on the size distribution at the beginning of the time step. Redistribution of the vapor phase occurs in a time that is small compared to the master time step; therefore, this redistribution is essentially decoupled from the other processes considered which justifies the use of a time parallel solution treatment.

Once redistribution of the vapor phase has been effected, its effect on the existing particle size distribution (in the volume in question) is calculated by assuming that each size class gains (or loses) mass in proportion to the rate of vapor transfer to (or from) that size class. Conservation of number for each size class then dictates redistribution between, in general, two new contiguous size classes, the number in each size class being determined by mass conservation.

At the end of a time step, the particle size distribution in each volume is re-evaluated over that time step to account for possible particle agglomeration, sources, and flow terms. The agglomeration algorithm has been excerpted from the QUICK aerosol behavior code⁽⁴⁾, which is based on a size discretization scheme.

The approximations inherent in this parallel treatment are minimized by relegating mass redistribution and conservation to Equation (1) except for redistribution due to radionuclide phase change. Agglomeration and particle evaporation/condensation serve only to modify the particle size distribution and therefore affect particle deposition indirectly through mass-distribution-averaged deposition velocities. Thus the aerosol aspect is solved (over a master time step) completely in parallel to Equation (1), using all sources, flow terms, and particle removal terms evaluated for each size class considered. The resultant distribution is used to evaluate average particle deposition terms for use in the master equation only. Similarly, reevaluation of the particle size distribution due to radionuclide phase change affects these average deposition terms only.

In addition to the time-dependent thermal hydraulic conditions and mass input rates by species, the TRAP-MELT 2 code requires input information on the initial particle size distribution of the source, the control volume geometry, and the physical properties of species (including deposition velocities on surface materials). The code provides output in terms of time- and location-dependent mass by species and state, as well as size distribution of suspended particulate material.

TRAP-MELT 2 Comparison with Other Codes

The TRAP-MELT 2 code can be compared with the RETAIN and RAFT codes in terms of differences in methods of analyzing sequences and in the mechanisms included. Table 1 summarizes the various mechanisms in each of the three codes.

Table 1. Phenomena Considered in Codes Predicting Fission Product Transport in Primary Systems

Phenomena/Parameters	CODE		
	TRAP-MELT 2.1	RETAIN	RAFT
<u>Phenomena:</u>			
Aerosol Agglomeration			
- Brownian	yes	yes	yes
- Gravitational	yes	yes	yes
- Turbulent	yes	no	yes
- Discretized Size Representation	yes	no	yes
Aerosol Deposition			
- Sedimentation	yes	yes	yes
- Thermophoresis	yes	yes	yes
- Laminar/diffusion	yes	yes	yes
- Turbulent/diffusion	yes	yes	no
- Turbulent/inertial	yes	yes	no
- Inertial (bends, obstacles)	no	no	no
- Electrostatic	no	no	no
- Diffusiophoresis	no	no	no
Vapor Deposition			
- Condensation/particles	yes	yes	yes
- Condensation/surfaces	yes	yes	yes
- Chemisorption/particles	no	no	no
- Chemisorption/surfaces	yes	no	no
- Nucleation	no	no	yes
Aerosol Resuspension	no	As Input	no
Channel Plugging	no	yes	no
Vapor Reevolution	yes	yes	yes
Combined Thermal Hydraulics	no	yes	no
Equil. Chem. Thermo.	no	no	yes
WASH 1400 f.p. Groups	yes	yes	By Element
Reverse Flow	yes	yes	no
<u>Parameters:</u>			
Input Particle Size (mean)	yes	yes	NA
Input Particle Density	yes	yes	NA
Gas Flow Rate w/time	yes	yes	yes
Gas Temperatures w/time	yes	yes	yes
Pressure	yes	yes	yes
Gas Composition	yes	yes	yes
Surface Temperatures w/time	yes	yes	yes
Multiple Surfaces/Volume	yes	no	no
Fission Product Mass Input w/time	yes	yes	yes
Mass Distribution among species w/time	yes	yes	yes
<u>Predictions (with time and location):</u>			
Particle composition	yes	yes	yes
Airborne vapor cone	yes	yes	yes
Condensed mass by species	yes	yes	yes
Sorbed mass by species	yes	yes	no
Released mass by species	yes	yes	yes
Particle Size Distribution			
Size Intervals	yes	no	yes
Mean Size	yes	yes	yes
Distribution Spread	yes	yes	yes

The RETAIN code⁽⁵⁾ predicts vapor and particulate transport in the RCS and was developed from an earlier version of the TRAP-MELT code and therefore is in many ways quite similar. There are, however, several major differences. The first major difference is that the RETAIN code employs the assumption that the size distribution for the aerosols remain log-normal. This assumption is reasonably accurate when low concentrations and low rates of gravitational agglomeration exist. However, for RCS conditions having high aerosol concentrations, the result is to overestimate the deposition rate.

The RETAIN code has the added feature of combining surface heatup from decay heating with the deposition melting. This allows for enhanced revaporization of fission products as a result of the increased deposit temperatures. This effect is expected to be quite important in later stages of an accident and in particular after reactor vessel melt-through.

The RAFT code⁽⁶⁾ predicts fission product transport and retention in the RCS and contains many of the same features as the TRAP-MELT and RETAIN codes, but also contains some significant modifications. Although it is limited to flow in one direction only, it tracks chemical changes in the vapor using chemical equilibrium thermodynamics and contains a model for nucleation of fission product vapor to form "new" particles. Theoretical predictions of nucleation rates are likely to be subject to large uncertainties and therefore a strong experimental validation of this portion of the code seems necessary if it is to be acceptable for accident analyses which represent a very complex situation. Conceptually, the inclusion of nucleation rates into the model is an attractive feature but its importance is still to be demonstrated.

IMPORTANT MECHANISMS

Among the mechanisms affecting fission product deposition within the reactor coolant system, the most important as identified from TRAP-MELT 2 calculations are aerosol agglomeration and sedimentation, and vapor condensation and reaction with surfaces. There are short periods of time during some accident sequences where thermophoretic deposition of aerosols is also very important. It should be recognized that materials nominally starting out as vapor, such as CsI or CsOH, are in many cases first condensed onto particles before being removed by aerosol deposition. Therefore a combination of vapor condensation with aerosol behavior becomes critical.

Thermal hydraulic conditions are predicted to have a major effect on deposition. Low gas flow rates lead to higher aerosol concentrations, high agglomeration rates, long residence times and hence, large fractions of the aerosol materials being deposited. The effect of temperature on condensation has been estimated to range from nearly zero to over 50 percent of CsOH deposited in the upper plenum as the upper plenum temperature was varied parametrically from high to low temperatures over a range expected to represent reasonable thermal hydraulic uncertainties.⁽¹⁾

Even though parametric studies have shown the importance of the various mechanisms included in the codes and the sensitivity to thermal hydraulic conditions is noted, questions still arise concerning mechanisms not currently modeled or modeled in a simplistic manner. Sensitivities to these additional mechanisms

are of possible importance and are often evaluated individually and external to the formalized computer codes. Some of these mechanisms and issues in question will be discussed below.

Revaporization by Decay Heating

A major issue being addressed theoretically at this time is the revaporization of previously deposited fission products by decay heating of the deposits. The conclusions are not yet finalized but some major observations can be made based on current progress. While airborne and after depositing fission products will continue to release decay energy, some of which will be deposited in the gas phase, some in the deposited layer, and some in the primary system structures. This distribution of energy seems to be treated in various ways by several investigators but the major concern has been the increasing temperature of the fission product layer which could lead to revaporization.

The various analyses to date have been consistent in suggesting the revaporization and subsequent redeposition of fission products within the primary system. The impact of this revaporization on release from the primary system is less clear. In some analyses heat loss from external primary system surfaces is sufficient to prevent significant loss from the primary system after a time of vaporization/condensation driven redistribution. Other analyses have not proceeded beyond the time of pressure vessel melt-through and hence long term effects are unknown.

It is the long term revaporization that seems crucial since many accident sequences lead to a predicted containment failure and extending the release from the primary system beyond containment failure could have significant effects on source term to the environment.

It is believed that considerable insight into the long term release will be forthcoming in the near future. There are both experimental and theoretical efforts under way at present.

Chemical Reactions

Chemical reactions have been generally ignored in analytical treatment of transport in primary systems. An exception is the RAFT code which performs chemical thermodynamic equilibrium calculations for the gas phase constituents. There are, however, a number of issues that must be resolved and are the subject of both theoretical and experimental studies.

The usual assumption made in primary system transport calculations is that CsI is the predominant iodine form, and it will transport as a vapor, condensing onto aerosols and surfaces at rates dictated by temperatures and flow conditions. There are several situations where this scheme may be altered. Based on theoretical considerations and supported by experiments, it is becoming evident that CsI may interact with boron compounds in the gas and condensed phases to produce molecular iodine. The kinetics of this process relative to subsequent iodine reactions must still be analyzed to evaluate what impact it may have on source terms to the environment.

Based on preliminary theoretical and experimental results, it appears possible to oxidize CsI at high temperatures such as in burning hydrogen to form molecular iodine. This appears to be most likely to occur in the containment or after melt-through of the primary system.

The chemical interactions of fission product vapors with surfaces has been studied more extensively from experimental approaches and have emphasized CsI, CsOH, and Te reactions with various structural materials. It appears that Te can be expected to react rapidly with structural materials while CsOH will react most effectively with trace components of such materials. The capacity of the surfaces for CsOH reaction seems to need more clarification to permit full utilization of the data.

In general, chemical interactions among fission products and between fission products and surfaces have the potential to alter volatilities. Either higher or lower vapor pressure products can be produced and the impact on condensation or revaporization could be substantial.

Resuspension of Aerosol Deposits

Resuspension of previously deposited aerosols on high pressure failure of the reactor vessel has been identified as a possible mechanism by which the fission product source to the containment could be increased. Reviews of this subject were undertaken as portions of both the QUEST and IDCOR programs. Resuspension can be considered for both liquid or slurry deposits and for dry deposits.

The resuspension of dry deposits could result during blowdown of the reactor pressure vessel if failure occurs at high pressure. The adhesion of particles depends on many factors including particle size, humidity, surface roughness, material characteristics (sticking tendencies), and depth of deposit. There is also a time factor with high gas velocities over the deposit surface being required for some length of time; the fraction resuspended increasing with time. The general conclusion is that for sequences with reactor vessel failure at high pressure, dry deposits would resuspend partially with the resuspended particle size being large enough to be of minor concern for containment transport.

For liquid deposits which appear more likely under primary system conditions (such as noted in the Marviken experiments with CsI, CsOH, and Te), resuspension could occur at flow discontinuities or from liquid surface instabilities. However, it appears unlikely that liquids would be resuspended to a significant extent during the blowdown period. Although some verification seems warranted, resuspension to form a readily transportable aerosol in the containment appears to be a minor effect.

Natural Circulation

Natural circulation flows can lead to exchanges among major portions of the primary system and control the extent of mixing within individual control volumes. The available transport codes are based on the assumption that

individual control volumes are well mixed. The TRAP-MELT 2 transport code maintains the capability for flow exchange in both directions between adjacent control volumes and hence could be matched with a thermal hydraulic prediction involving recycling flows. Efforts supported by both the U.S. Nuclear Regulatory Commission and the Electric Power Research Institute are directed toward more comprehensive descriptions of thermal hydraulic conditions including natural circulation.

The importance of natural circulation is expected to be dependent on accident conditions. Preliminary calculations have indicated that for the upper plenum, a hot leg break leads to unmixed conditions while a transient accident gives a well mixed situation. Analyses extended to include more portions of the primary system suggest larger circulation patterns among major components. The implication of these results for primary system transport and deposition is that a finer nodalization of control volumes may be needed and somewhat revised definitions of control volumes and their interchanges would be required.

CONCLUSIONS

The general conclusions regarding fission product transport and deposition in the primary system are:

- (1) There appears to be a solid basis for theoretical predictions.
- (2) The predictive codes will require improvement to consider additional chemical and physical effects.
- (3) Decay heating of deposits and chemical changes and interactions among fission products and with surfaces may have a pronounced effect on release from the primary system, particularly over long time periods.
- (4) Resuspension of deposits within the primary system is probably not a major source of transportable aerosol.
- (5) Natural circulation is important in some accident sequences, may have a significant effect on thermal conditions, and may lead to revisions in transport codes.

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30

AEROSOL BEHAVIOR IN A STEAM-AIR ENVIRONMENT*

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ABSTRACT

The behavior of aerosols assumed to be characteristic of those generated during light water reactor (LWR) accident sequences and released into containment is being studied in the Nuclear Safety Pilot Plant (NSPP). Observations on the behavior of U_3O_8 aerosol, Fe_2O_3 aerosol, concrete aerosol, and various mixtures of these aerosols in a dry air environment [relative humidity (RH) less than 20%] and in a steam-air environment [relative humidity (RH) approximately 100%] within the NSPP vessel (38.3 m³ volume) are reported. The primary experimental variables are aerosol mass concentration and aerosol mass ratios.

Under dry conditions the three individual aerosols and mixtures of these aerosols behave differently with regard to rate of removal from the vessel atmosphere. The aerosols are agglomerated in the form of branched chains; the aerodynamic mass median diameter (AMMD) of the U_3O_8 , Fe_2O_3 and mixed U_3O_8 - Fe_2O_3 aerosols ranged between 1.5 and 3 μm while that of the concrete aerosol was about 1 μm .

Comparison of the behavior of these same aerosols in steam-air with that observed in dry air reveals that a steam-air environment, which would be present in LWR containment during and following an accident, causes the U_3O_8 , the Fe_2O_3 , and mixed U_3O_8 - Fe_2O_3 aerosols to behave in a manner different from that observed in a dry atmosphere; the primary effect is an enhanced rate of removal of the aerosol from the vessel atmosphere. Concrete aerosols were observed to behave differently. Based upon results to date, the presence of steam does not have a significant effect on the removal rate of a concrete aerosol. Electron microscopy showed the agglomerated U_3O_8 , Fe_2O_3 , and mixed U_3O_8 - Fe_2O_3 aerosols to be in the form of spherical clumps of particles differing from the intermingled branched chains observed in the dry air tests; the AMMD was estimated to be in the range of 1 to 2 μm . Steam seemed to have a lesser influence on the physical shape of the concrete aerosol with the shape being intermediate between branched chain and spherical clumps.

The enhanced rate of removal of the U_3O_8 , the Fe_2O_3 , and the mixed U_3O_8 - Fe_2O_3 aerosols from the atmosphere of the NSPP vessel by steam is probably caused by the change in aerosol shape and the condensation of steam on the aerosol surfaces combining to increase the effect of gravitational settling. The apparent lack of an effect by steam on the removal rate of concrete aerosol could result from a differing physical/chemical response of the surfaces of this aerosol to condensing steam.

INTRODUCTION

The behavior of aerosols assumed to be characteristic of those generated during light water reactor (LWR) accident sequences and released into containment is being studied in the Nuclear Safety Pilot Plant (NSPP) which is located at the Oak Ridge National Laboratory (ORNL). This project, which is part of the ORNL Aerosol Release and Transport (ART) Program, is sponsored by the Division of Accident Evaluation, Nuclear Regulatory Commission, and the purpose is to provide experimental qualification for LWR aerosol behavior codes under development.

The program plan for the NSPP aerosol project provides for the study of the behavior, within containment, of simulated LWR accident aerosols emanating from fuel, reactor core structural materials, and from concrete-molten core materials interactions. The aerodynamic behavior of each of these aerosols was studied individually to establish its characteristics; current experiments involve mixtures of these aerosols to establish their interaction and collective behavior within containment. Tests have been conducted with U_3O_8 aerosols, Fe_2O_3 aerosols, and concrete aerosols in an environment of either dry air [relative humidity (RH) less than 20%] or steam-air [relative humidity (RH) approximately 100%] with aerosol mass concentration being the primary experimental variable. Experiments are underway involving mixtures of these aerosols, and, to date, the test aerosol mixtures have been Fe_2O_3 + concrete and Fe_2O_3 + U_3O_8 ; in these tests the primary experimental variables have been aerosol mass concentration and aerosol mass ratio.

EXPERIMENTAL

The NSPP facility, shown schematically in Fig. 1, includes a test containment vessel, aerosol generating equipment, analytical sampling and system parameter measuring equipment, and an in-vessel liquid spray decontamination system. The NSPP vessel is a stainless steel cylinder with dished ends having a diameter of 3 m, a total height of 5.5 m, and a volume of 38.3 m^3 . The floor area is 7.7 m^2 and the internal surface area (including top, bottom, and structural items) is 68.9 m^2 . The equipment for the measurement of aerosol parameters includes filter samplers for measuring the aerosol mass concentration, coupon samplers for aerosol fallout and plateout measurement, cascade impactors and a centrifuge sampler for determining the aerodynamic particle size distribution of the aerosol, and devices for collecting samples for electron microscopy. System parameters measured are moisture content of the vessel atmosphere, steam condensation rates on the vessel wall, temperature of vessel atmosphere, temperature gradients near the wall, and vessel pressure.

For the dry aerosol tests the vessel atmosphere was dry air (RH <20%) and the temperature and pressure were slightly above ambient. The slight elevations in temperature and pressure result from the heat produced and gases injected by the plasma torch aerosol generator.

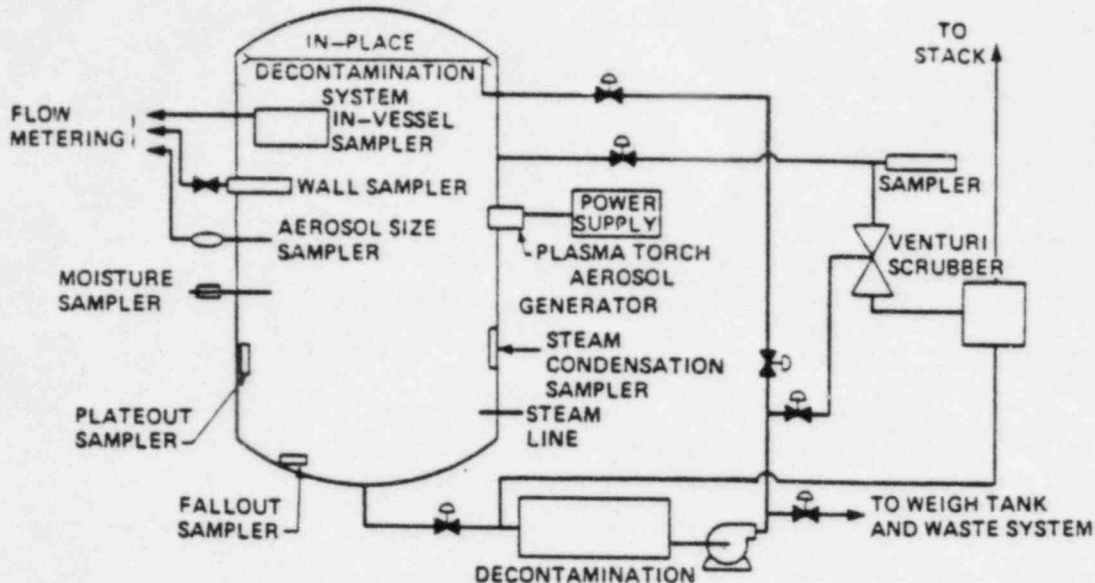


Figure 1. Diagram of the NSPP Facility.

The steam-air aerosol tests were conducted under quasi-steady-state steam conditions. The test atmosphere was prepared by injecting steam into the vessel (initially at subatmospheric pressure) to form a steam-air mixture at elevated temperature and pressure (around 380 K and at an absolute pressure of about 0.2 MPa); upon achieving this condition the rate of steam injection was reduced and the accumulated steam condensate removed from the vessel. The test aerosol was then introduced and steam injection was continued for six hours at a low rate to balance steam losses by wall condensation and assure maintenance of the quasi-steady-state conditions.

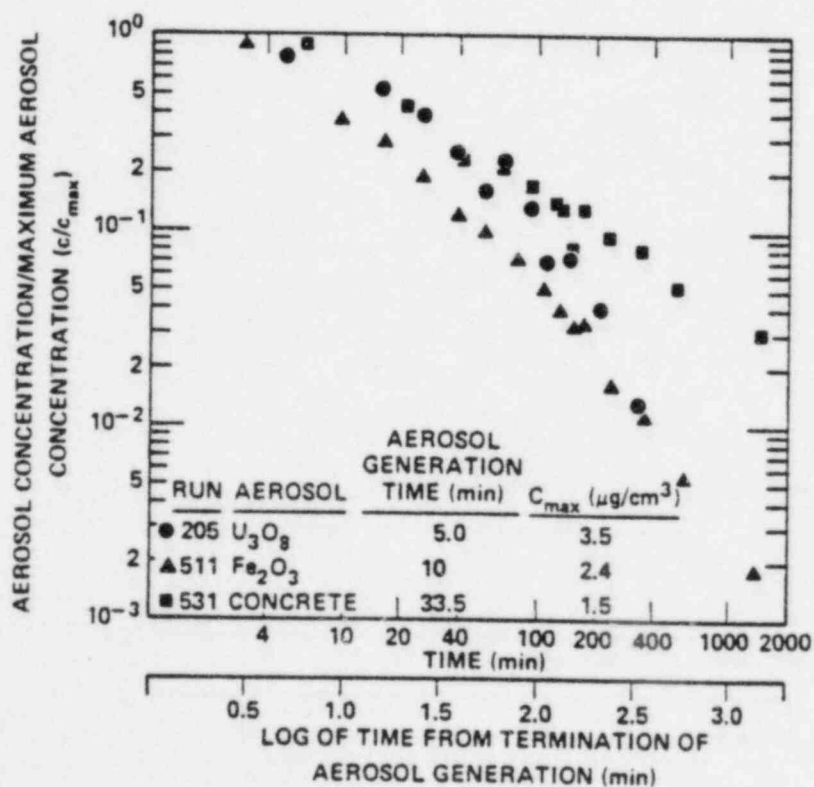
Single-Component Aerosol Tests

A number of single-component aerosol tests have been conducted under both dry air and steam-air test environments. Table I lists these tests; results from these tests have been reported [1, 2, 3].

Under dry conditions the three aerosols, U_3O_8 , Fe_2O_3 , and concrete, behave in a different manner with regard to rate of removal (decrease in aerosol mass concentration) from the vessel atmosphere. Figure 2 compares the behavior of the three aerosols. (Note that the aerosol mass concentration is normalized with respect to maximum concentration and that time is measured from the time of termination of aerosol generation for the purpose of comparison.) Scanning electron microphotography (SEM) shows the aerosols to be agglomerated in the form of branched-chains (Fig. 3). Particle size measurements by cascade impactors and spiral centrifuges indicated that the aerodynamic mass median diameter (AMMD) [4] of the U_3O_8 and Fe_2O_3 aerosols ranged between 1.5 and 3 μm while that of the concrete aerosol was about 1 μm , or less. Based upon

Table I. Details of single-component aerosol tests

Test Nos.	Aerosol	No. of tests	Test environment	Aerosol conc. range ($\mu\text{g}/\text{cm}^3$)
201-7, 209	U_3O_8	8	Air (dry)	0.05-9.0
208, 210	U_3O_8	2	Air (moist)	7.1, 12.5
401-4, 406-7	U_3O_8	6	Air-steam	5.8-28.0
511	Fe_2O_3	1	Air (dry)	2.4
501-2	Fe_2O_3	5	Air-steam	1.0-8.5
531	Concrete	1	Air (dry)	1.5
521-2	Concrete	2	Air-steam	1.1, 1.5

Figure 2. Behavior of Various Single-Component Aerosols in a Dry Air Environment ($\text{RH} < 20\%$).

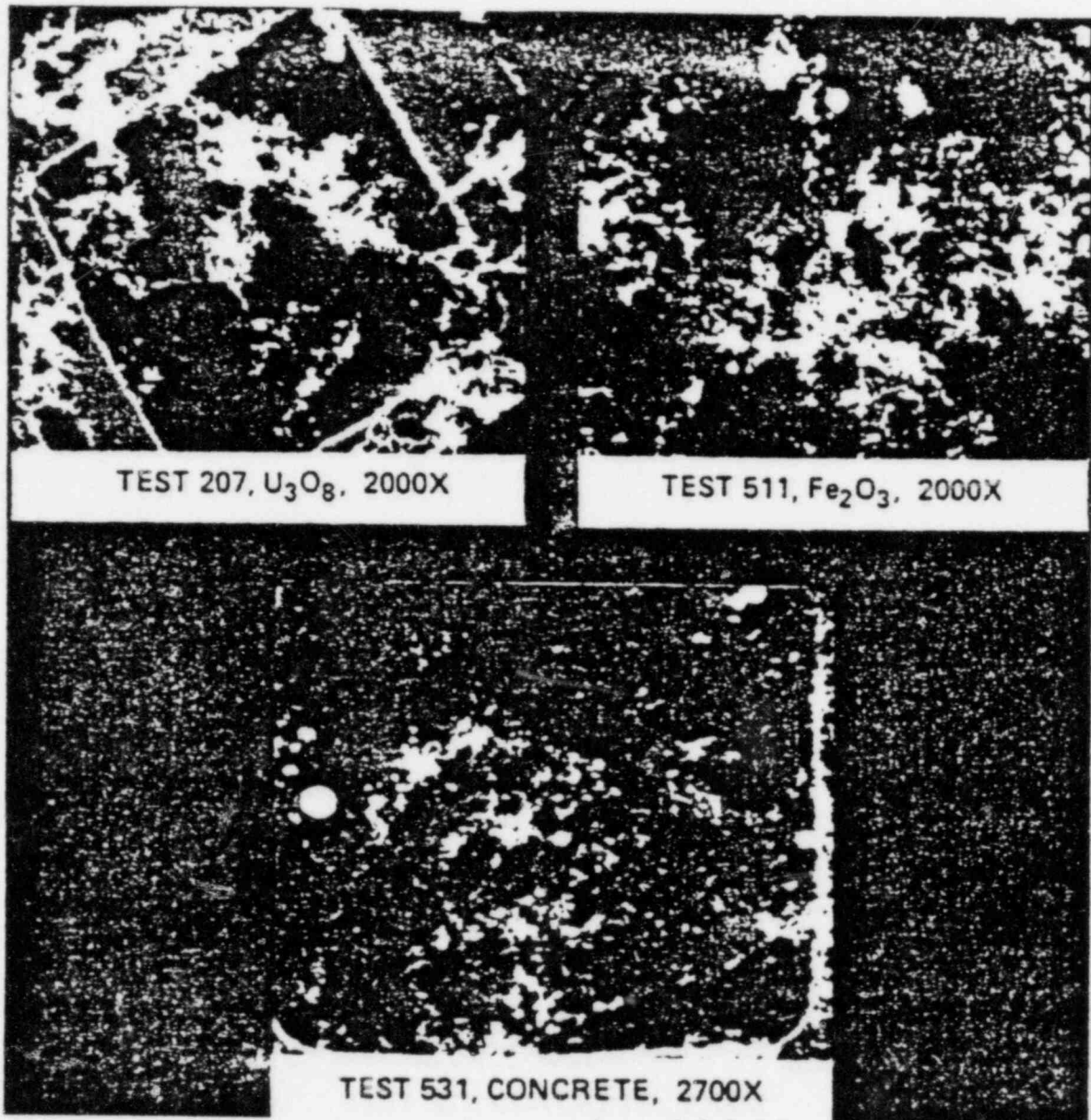


Figure 3. SEM Photographs Illustrating Typical Appearance of Chain-Agglomerate Aerosols in a Dry Air Environment (RH <20%).

the results from these tests under dry conditions, it has been observed that these aerosols have similar sizes and shapes but act aerodynamically in a different fashion.

The presence of steam in the test environment causes a change in both the aerodynamic behavior and the physical shape of these aerosols. The aerodynamic behavior of the aerosols is compared in Fig. 4. The most obvious effect of steam is an enhanced rate of aerosol removal from the vessel atmosphere in the case of U₃O₈ and Fe₂O₃ aerosols. For example, in Fig. 2 under dry conditions, the time required for 99% of

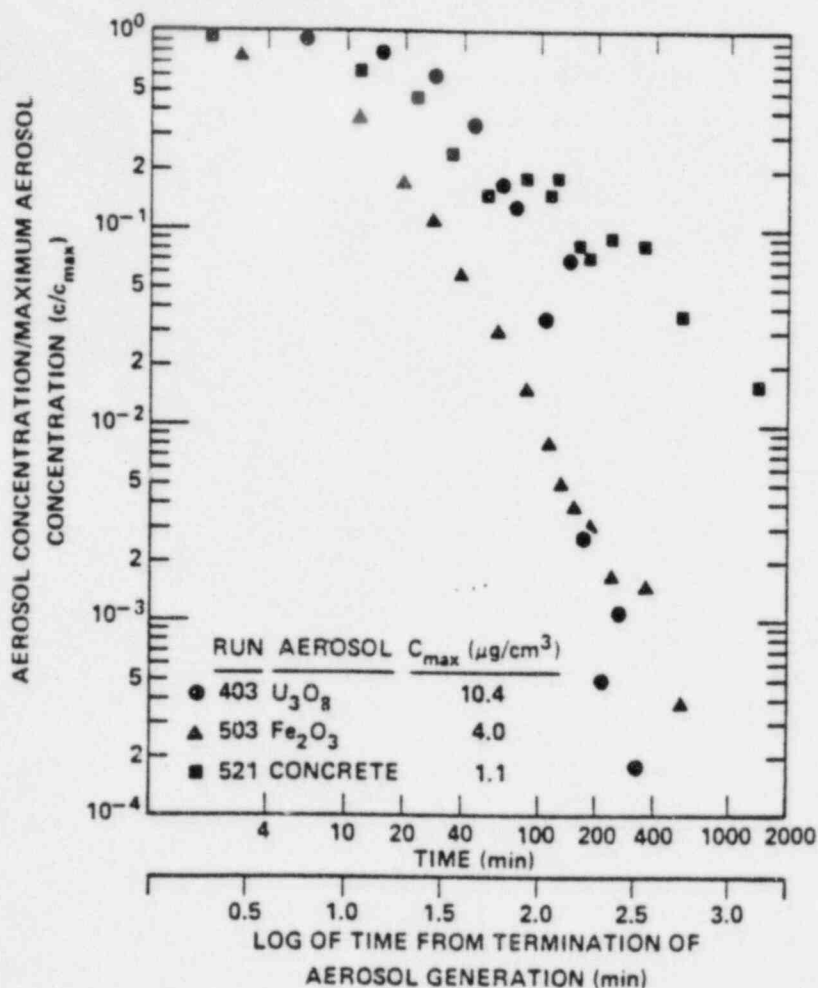


Figure 4. Behavior of Various Single-Component Aerosols in a Steam-Air Environment (RH ~100%).

the Fe_2O_3 aerosol to disappear from the vessel atmosphere is about 350 min.; under steam-air conditions this time is about 100 min. A similar comparison can be made for U_3O_8 aerosol. The shape of these two aerosols is changed from chain-agglomerate to almost spherical by the presence of steam as illustrated in Fig. 5 for U_3O_8 . The AMMD for the U_3O_8 or Fe_2O_3 aerosols in steam range from about 1 to 2 μm .

Concrete aerosol does not seem to be affected by the presence of steam in the same manner as U_3O_8 or Fe_2O_3 aerosol. This lack of influence is illustrated in Fig. 6 where the rates of removal of concrete aerosol under dry and under steam-air conditions are compared. This aerosol was generated by passing powdered limestone-aggregate concrete through the plasma torch aerosol generator. The concrete aerosol is not a simple, single-component, aerosol such as U_3O_8 or Fe_2O_3 ; it is actually a complex mixture of Al_2O_3 , SiO_2 , CaO , MgO , Fe_2O_3 , and various silicates with Al, Ca, Mg, and Fe as the cations. Steam also affects the physical shape of concrete aerosols (possibly to a slightly lesser degree than for U_3O_8 or Fe_2O_3) producing some spherical agglomerates.

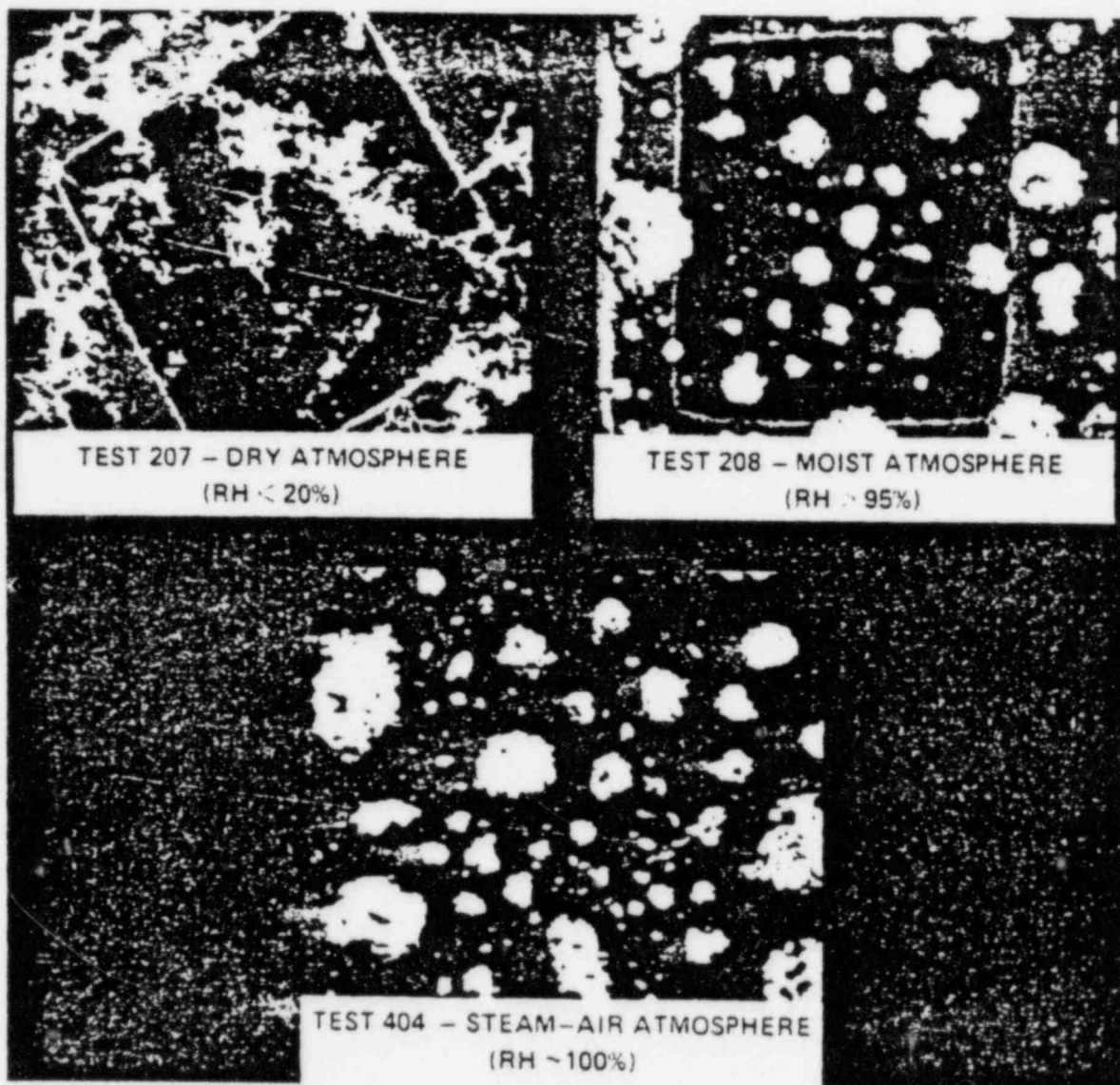


Figure 5. SEM Photographs Illustrating Influence of Moisture/Steam on Physical Shape of U₃O₈ Aerosol.

Figure 7 contains scanning electron microphotographs of a concrete aerosol in a dry air and in a steam-air atmosphere.

Multi-Component Aerosol Tests

Recent activities in the NSPP involve the study of the behavior of multi-component (mixed) aerosols in both dry air and steam-air environments. Details of these tests are contained in Table II. The first mixed aerosol to be studied in detail is U₃O₈ + Fe₂O₃. This mixture simulates those aerosols emanating from molten fuel and molten-core support and structural materials. Experimental procedures are essentially

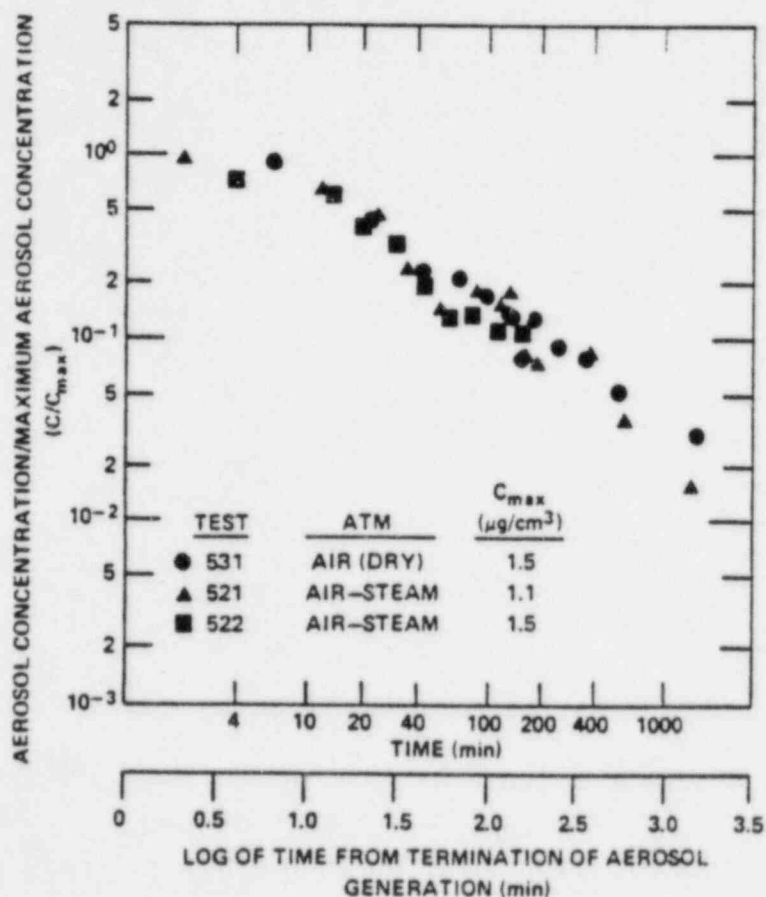
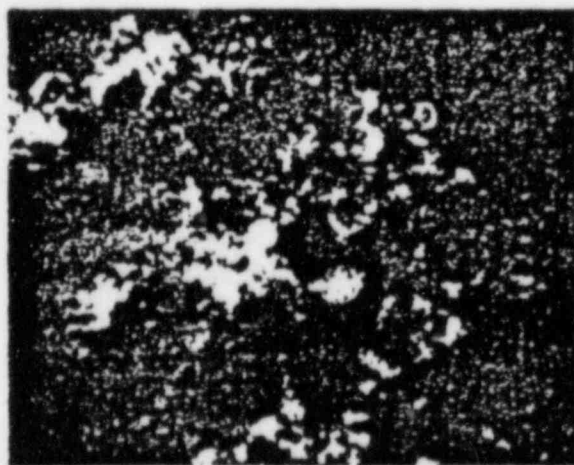
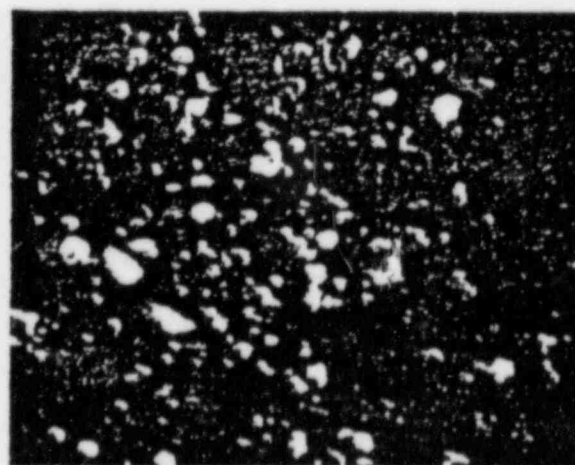


Figure 6. Behavior of Concrete Aerosol in a Dry Air (RH <20%) and a Steam-Air (RH ~100%) Environment.



TEST 531 - DRY ATMOSPHERE
(RH < 20%)
6300X MAG.



TEST 052 - STEAM-AIR ATMOSPHERE
(RH ~ 100%)
9000X MAG.

Figure 7. SEM Photographs Illustrating Influence of Steam-Air on Physical Shape of Concrete Aerosol.

Table II. Details of multi-component aerosol tests

Test No.	Mixed aerosol	Test environment	Max. aerosol conc. ($\mu\text{g}/\text{cm}^3$)	Mass ratio ($\text{Fe}_2\text{O}_3/\text{U}_3\text{O}_8$)
601	Concrete + Fe_2O_3	Air-steam	5.5 2.5	-
611	U_3O_8 + Fe_2O_3	Air-steam	4.0 5.5	1.4/1
612	U_3O_8 + Fe_2O_3	Air-steam	1.8 0.5	0.3/1
613	U_3O_8 + Fe_2O_3	Air-steam	0.7 6.8	9.7/1
631	U_3O_8 + Fe_2O_3	Air (dry)	1.7 1.2	0.7/1

the same as for the single-component aerosol tests. The principal difference is in aerosol generation; the U_3O_8 and Fe_2O_3 aerosols are produced with separate plasma torch generators and allowed to mix within the vessel.

Four mixed aerosol experiments involving various mixtures of Fe_2O_3 and U_3O_8 aerosols have been completed; three were conducted in a steam-air environment and one in a dry air (RH <20%) environment. The behavior of the mixed aerosol (Fe_2O_3 + U_3O_8) in a steam-air environment has been similar in the three experiments conducted, although the mass ratio of Fe_2O_3 to U_3O_8 has been different in each case. The aerosol mass fraction airborne (C/C_{max}) as a function of time after termination of aerosol generation is illustrated in Fig. 8 for these experiments. Although the rate of aerosol removal during the first 30 min is somewhat larger in Exps. 611 and 613 as compared to Exp. 612, the time required for 99% removal of aerosol mass from the volume of the vessel is about 60 min in all three experiments. SEM photographs of the mixed aerosol showed almost spherical clumps of aerosol in each case. The AMMD of the mixed aerosol in all cases was in the 1 to 1.7- μm range.

To illustrate the effect of steam on the behavior of the mixed aerosol, the results from experiment 631 are compared with those of Nos. 611-613 in Fig. 8. Under dry air conditions, the mixed aerosol tends to remain airborne longer than under steam-air conditions. Note that the time required for 99% of this aerosol to be removed from the vessel is about 400 min as compared with 60 min for the aerosol in the steam-air environment. SEM photographs show the aerosol to be in the form of chain-agglomerates (also observed in previous experiments with Fe_2O_3 or

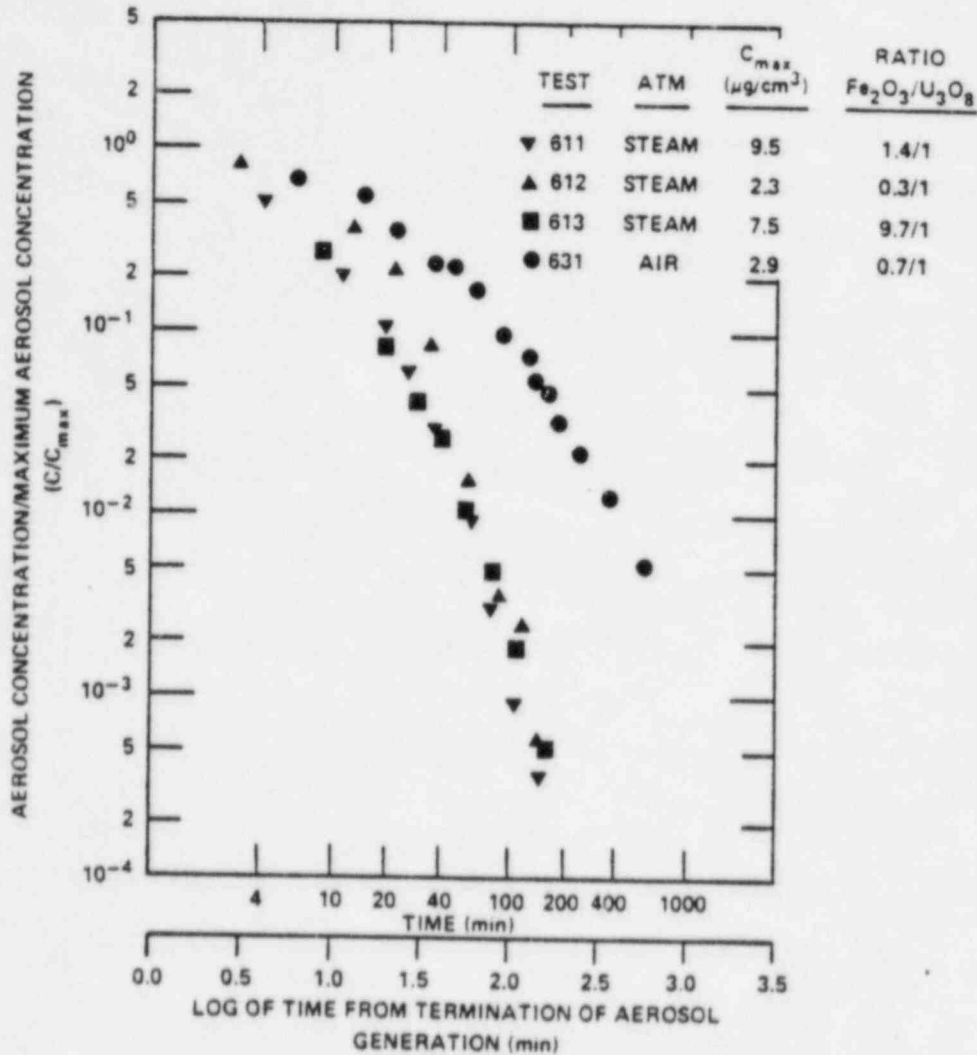


Figure 8. Comparison of Behavior of Multi-Component Aerosol $\text{Fe}_2\text{O}_3 + \text{U}_3\text{O}_8$ in Steam-Air (RH ~100%) and Dry Air (RH <20%) Environments.

U_3O_8 aerosol in dry air) rather than in spherical clumps as in Nos. 611-613. The AMMD for the mixed aerosol is slightly larger in the dry atmosphere with a value as large as $2.7 \mu\text{m}$ being observed.

It appears, based upon limited data, that the influence of one aerosol component on the other, in a mixed aerosol, can be significant. The behavior of the mixed $\text{Fe}_2\text{O}_3\text{-U}_3\text{O}_8$ aerosol is more like that of Fe_2O_3 aerosol than U_3O_8 aerosol. Data are available which permit a comparison of the influence of concrete aerosol and U_3O_8 aerosol in a mixture with Fe_2O_3 aerosol. Figure 9 compares the behavior of a $\text{Fe}_2\text{O}_3 + \text{concrete}$ aerosol with a $\text{Fe}_2\text{O}_3 + \text{U}_3\text{O}_8$ aerosol in a steam-air environment. $\text{Fe}_2\text{O}_3 + \text{concrete}$ aerosol at a mass ratio of 0.45 to 1 (Fe_2O_3 to concrete) behaves more like a concrete aerosol; $\text{Fe}_2\text{O}_3 + \text{U}_3\text{O}_8$ aerosol at

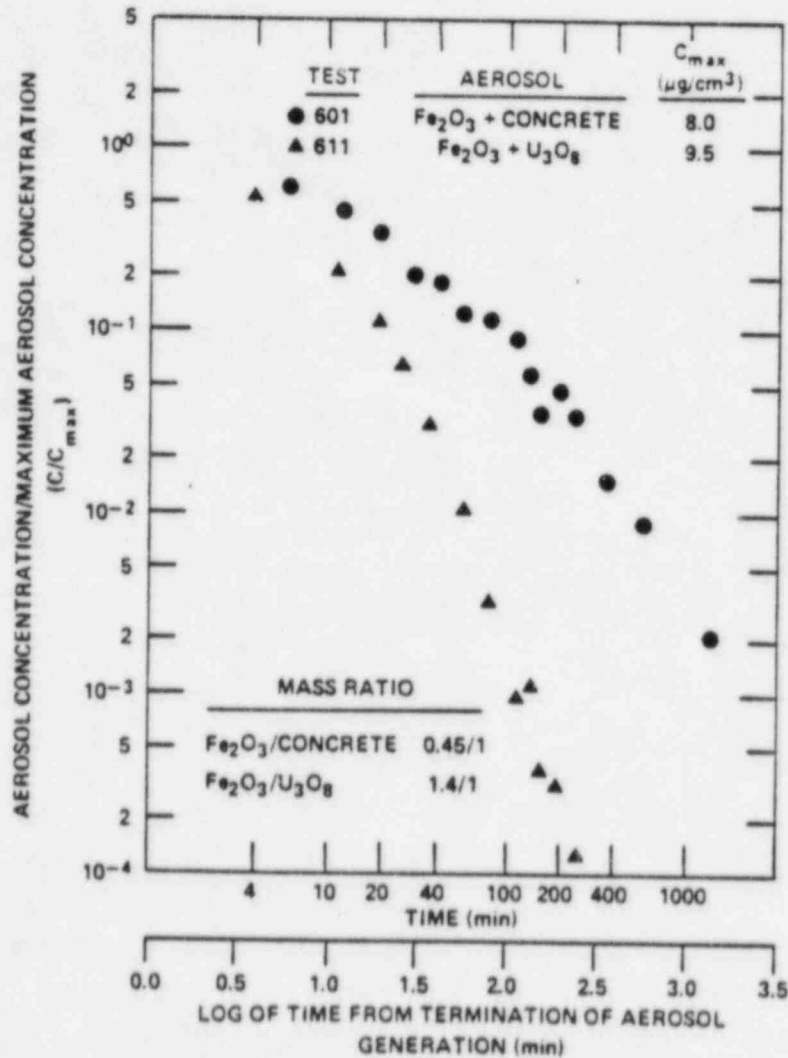


Figure 9. Comparison of Behavior of Multi-Component Aerosols in a Steam-Air Environment (RH ~100%).

a mass ratio of 1.4 to 1 (Fe_2O_3 to U_3O_8) behaves more like a Fe_2O_3 aerosol. Future tests on mixed aerosols will permit a more definitive examination of the influence of one component on another in mixed aerosols.

SUMMARY

General statements may be made on the behavior of single-component and multi-component aerosols in the NSPP vessel. The removal processes for U_3O_8 , Fe_2O_3 , and $\text{U}_3\text{O}_8 + \text{Fe}_2\text{O}_3$ aerosols are enhanced in a steam-air atmosphere. Steam-air seems to have little effect on removal of concrete aerosol or $\text{Fe}_2\text{O}_3 + \text{concrete}$ aerosol from the vessel atmosphere. A steam-air environment causes a change in aerosol shape from chain-agglomerate to basically spherical for U_3O_8 , Fe_2O_3 , and $\text{U}_3\text{O}_8 + \text{Fe}_2\text{O}_3$

aerosol; for concrete and Fe_2O_3 + concrete aerosol the change in aerosol shape is from chain-agglomerate to partially spherical. The mass ratio, as well as the identity, of the individual components of a multi-component aerosol seems to have an observable influence on the resultant behavior of these aerosols in steam.

The enhanced rate of removal of the U_3O_8 , the Fe_2O_3 , and the mixed U_3O_8 + Fe_2O_3 aerosols from the atmosphere of the NSPP vessel by steam-air is probably caused by the change in aerosol shape and the condensation of steam on the aerosol surfaces combining to increase the effect of gravitational settling. The apparent lack of an effect by steam-air on the removal rate of concrete aerosol could result from a differing physical/chemical response of the surfaces of this aerosol to condensing steam.

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