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DIRECT IN-VESSEL APPLICATIONS EXPERIMENTS
AT HARVARD AIR CLEANING LABORATORY

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ABSTRACT

Prototypes of direct in-vessel emergency air cleaning systems were evaluated for possible application in an LMFBR containment vessel after a hypothetical core disruptive accident. These were turbulence enhanced sedimentation, powder dispersal scavenging, acoustic agglomeration enhanced sedimentation, and combinations of turbulence with powder dispersal. The effect of turbulent agglomeration in enhancing the sedimentation of a sodium pool fire aerosol was experimentally demonstrated in a 90 m³ test chamber, 4 meters high. Two hour dose reduction factors (DRF(2 hr)) from 7 to 56 were achieved in the 4 meter high chamber using fan-induced turbulent agglomeration on aerosols that varied in initial mass concentration from 1 to 12 gm/m³. In the same chamber, a prototype limestone powder dispersal scavenging system was tested and achieved DRF(2 hr)'s up to 10. The beneficial effect of combining turbulence with powder dispersal in a single system was demonstrated in a test which yielded a DRF(2 hr) of 20. This was greater than for either mechanism separately applied when initial aerosol mass concentration was 2 gm/m³.

Acoustic agglomeration of sodium pool fire aerosols was tested in a smaller 0.65 m³, 75 cm high settling chamber, using an electronic siren which produced a sound pressure level of 145 dB in the reverberant chamber. The DRF(2 hr) in the small chamber with the siren operating was found to be from 17 to 31, 2 to 3 times greater than the DRF(2 hr) for unperturbed settling. Pulse-jet engines were found to be unsuitable for generating high sound levels for this application. Scaling each of the systems tested to a 30 m high containment vessel indicated no reason why one or more of the systems investigated could not be applied successfully as an emergency air cleaning system.

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INTRODUCTION

The liquid metal fast breeder power reactor (LMFBR), now under intensive development, is designed to greatly extend the use of uranium fuel by breeding fissionable plutonium from the relatively abundant U^{238} uranium isotope. The necessary very high reactor core power density and the need for a low neutron absorption cross section coolant make liquid sodium metal ideal for this application. A core disruptive accident (CDA) which might leak thousands of pounds of molten sodium into the reactor vessel containment building is considered to be a very low probability occurrence, but one for which regulatory policy demands an accounting.

The sodium in the primary coolant loop of the LMFBR is made radioactive by the intense neutron flux in the core. Escaped coolant, reacting in the containment atmosphere, to the various oxides, hydroxides, and carbonates of sodium is, therefore, a radiological hazard that demands attention. Radioactive nuclides present in an aerosol cloud resulting from an accident, particularly plutonium, would pose health hazards to the surrounding environment should they leak from the containment building. The intact containment leakage rate is, for some designs not more than 0.1% of the total contained atmosphere (containment vessel volume = $7 \times 10^4 \text{ m}^3$) per 24 hour period. Natural settling of the aerosol in the containment vessel would reduce the mass concentration exponentially. The amount of radiological material escaping to the external environment could be dramatically reduced with the application of an emergency air cleaning system (EACS) capable of rapidly reducing concentration of hazardous aerosol particles in the containment building atmosphere.

Three modes of operation of an EACS have been considered: gas recirculation within containment through a gas cleaning device, gas purged outside containment through a gas cleaning device, and direct in-vessel reduction of airborne concentrations by systems which treat the entire containment vessel atmosphere at once.(1) Although very rapid reductions in aerosol concentration could be achieved using gas recirculation and gas purging, this would require extremely large filtration and air flow systems. Such systems would call for a large capital investment, require large amounts of power (which might not be available during a major reactor accident), and occupy a large fraction of the containment volume. Useful direct in-vessel systems, on the other hand, would have a potential for rapid reduction of aerosol concentration with modest energy input and small space requirements.

A spectrum of direct in-vessel application (DIVA) methods has been proposed by HEDL.(1) These include powder dispersal scavenging, liquid spray scavenging, foam encapsulation, acoustic agglomeration enhanced sedimentation, and electrostatic precipitation. Combinations of the latter methods, whether concurrent or intandem, have also been proposed. The HACL research program selected several of the most promising concepts for experimental evaluation by prototype tests with sodium pool fire aerosols. The experiments that were conducted at HACL, were not intended to be exhaustive but to simulate the sodium fire conditions which might be experienced in a real accident. Therefore, the methods selected for testing were not intended to imply rejection of other direct application methods.

We selected induced turbulent agglomeration, a method not previously proposed, for initial testing in our 4 meter high, 90 m³ test chamber. Later we explored powder dispersal scavenging using inert dusts and acoustic agglomeration of sodium aerosols. Combinations of turbulent agglomeration and powder dispersal scavenging to produce an advantageous synergistic effect, were also investigated. When evaluating these prototype systems, the focus was on the "two-hour dose reduction factor" figure of merit proposed by some investigators.(2) The "dose reduction factor" achieved in a two hour period following the start of an air cleaning system (at t=0) is:

$$\text{DRF}(2 \text{ hr}) = \frac{C_0 t_*}{\int_0^{t_*} C(t) dt} \quad \text{for } t_* = 2 \text{ hours} \quad (1)$$

where C_0 is the initial aerosol mass concentration and $C(t)$ is the mass concentration time history of the aerosol affected by the air cleaning system. A dose reduction factor of 10 for a two-hour period is considered highly desirable for containment vessels that are approximately 30 m high. For the present investigation, theoretical models were required for extrapolating performance of prototype DIVA systems to containment vessel dimensions.

EXPERIMENTAL METHODS AND REFERENCE AEROSOL CHARACTERISTICS

1. Aerosol Generation and Measurement

To evaluate the effectiveness of the direct in-vessel methods under investigation, it was necessary to establish as a baseline the aerosol sedimentation characteristics of an unperturbed cloud generated by a sodium pool fire. The baseline test in our air-filled, 4 m high, 90 m³ rectangular chamber involved melting and burning one pound of sodium inside an electrically heated, insulated steel pot. The relative humidity of the air-filled chamber at the start of the sodium fire was approximately 20% for all baseline fires. Sodium combustion in a normal atmosphere results in a very dense, white aerosol which in minutes fills the chamber. The mass concentration decay with time, the baseline property of most importance, was obtained with open-face absolute filter samples taken near the mid-chamber height. Generally, samples were taken over 3 minute intervals at 18.4 l/m.

The baseline sodium aerosol generation and decay characteristics shown in Figure 1, were derived from eight sodium fires. Natural decay by gravitational settling gave a dose reduction factor in 2 hours of 1.45 when measured from the peak concentration point which occurs at approximately 25 minutes after the initiation of burning. This average DRF(2-hr) value corresponds closely to the decay characteristics of stirred settling of a spherical 4 μ m monodisperse aerosol particle with a true density of 1.0 gm/cm³. Scaling the settling of this aerosol for a 30 m high chamber by assuming stirred settling for a unit density 4 μ m monodisperse aerosol, the equivalent DRF(2-hr) would be 1.05.

The mass median diameter (MMD) and geometric standard duration (GSD) of the baseline aerosol cloud were determined by eight-stage Andersen impactor tests and found to be nearly constant near 3 μm starting 40 minutes after initiation of the fire, as shown in Figure 2. This behavior suggests that there may be processes present that are analogous to those involved in the "self-preserving" aerosol size distribution postulated by Lai, et al.(3) The sample taken at 20 minutes indicates a MMD close to 2 μm , showing that the aerosol is still rapidly agglomerating. These size measurements are consistent with results from sodium pool fires conducted by Atomics International(4) (their Figures A.1.10 and A.1.11). Their GSD over the period covered in the measurements is in the range 1.4 to 1.6 and their MMD is slightly over 3 μm for several hundred minutes.

The baseline sodium aerosol was sampled more extensively with an aerosol centrifuge(5), which fractionates particles according to aerodynamic diameter. Centrifuge data from a number of runs gave an average MMD of 1.85 μm and an average GSD of 1.65.(6) The lower MMD determined by centrifuge versus that obtained from the eight-stage Andersen impactor might be due to size selective losses at the aerosol inlet. Aerodynamic size calibration of the centrifuge was performed with polystyrene latex spheres of known size and density. These give reliable calibration values over the size range of interest.

During the baseline tests the measured ratio of chamber floor to chamber wall deposits (mass per unit area) was in the range of 10 to 80. The large spread was caused, in part, by uncertainty in the measurement of the very low wall deposits collected on filter papers.

2. Chemical Composition of the Aerosol

Chemical and physical tests were performed to determine the composition of some of the filter samples. Sodium deposit measurements by atomic absorption spectrophotometry indicated high ratios of total sample mass to sodium mass. These ratios were often much larger than would be associated with a nonhydrated sodium compound. The conclusion was that most of the sodium compounds on the filter sample contain water of hydration.

The data in the form of the ratio of total mass to sodium mass, R , are plotted in Figure 3 as a function of time. As shown in Figure 3, there is no apparent trend in the value of R with time. The average value of R from the 24 measurements is 2.95 with a standard deviation of 0.65. A check of the atomic absorption measurements was made by determining the value of R by wet chemical analysis of the filter deposits rather than by atomic absorption analysis for sodium. This procedure gave values of R from 2.84 to 2.93 for times ranging from 28 minutes to 268 minutes, in close agreement with the atomic absorption values.

Quantitative chemical analysis for peak mass concentration $\leq 2 \text{ gm/m}^3$ indicates that the particles are completely converted to carbonate with less than 1% of the oxide or hydroxide remaining. This is consistent with the rates of the reaction for oxide to hydroxide to carbonate formation described by Clough and Garland.(7) Peak aerosol

mass concentrations during the chemical evaluation tests were from 1.6 to 3.6 gm/m³. The air-filled chamber had sufficient CO₂ present initially to produce an airborne Na₂CO₃ concentration of 1.3 gm/m³, assuming 100% utilization of CO₂, with the rest of the mass loading (i.e., above 1.3 gm/m³) being water of hydration and excess water adsorbed by the aerosol particles. The peak concentrations and total mass to sodium mass ratio data together indicate mixtures of carbonate compounds and their hydrates.

The reaction of carbonate formation from the initial sodium oxide and sodium superoxide combustion product is:

Theoretical analyses performed by other investigators(7) show reaction rates of only seconds for forming Na₂CO₃ from the hydroxide primary compound--a fact which is consistent with measured chemical composition data.

TURBULENT AGGLOMERATION TESTS

The first full chamber tests of a direct precipitation method involved the use of turbulence to enhance agglomeration and speed sedimentation. A large (3000 cfm) centrifugal recirculating blower placed in the center of the chamber produced DRF(2-hr) values in the range 4.2 to 8.2 when it was started shortly after the peak aerosol concentration was reached. The time decay history of the first five tests is presented in Figure 4. In an effort to separate the relative effects of centrifugal air cleaning in the blower shroud from the effect of turbulence enhanced agglomeration and sedimentation, the blower was run with and without the fan shroud in place. Operation with the shroud off takes away a major surface for centrifugal deposition, though some cleaning credit must still be taken because of particle impaction on the fan blades. Removing the shroud did not greatly change the DRF(2-hr) from shroud-on performance, as can be seen in Figure 4.

Substantially higher DRF(2-hr)'s were obtained with turbulent agglomeration when the peak mass concentration was increased from the baseline peak value (~2 gm/m³). Higher concentrations were generated by melting larger quantities of sodium and by faster burning rates induced by blowing an air jet against the sodium pool surface. Figure 5 shows the mass concentration profiles of two of these tests which resulted in a DRF(2-hr) of 28 and 43, respectively. Test R-21 was conducted with air-augmented burning of 1 lb. sodium whereas test R-22 involved air-augmented burning of 3 lbs. of sodium. For both of these tests, blower turbulence began when mass concentrations in the chamber were above the baseline level. This confirms that higher initial particle number concentrations accelerated agglomeration in the expected fashion. Figure 5 shows another run with DRF(2-hr) greater than 8 in which turbulence was initiated after the mass concentration had peaked and then dropped back to 2 gm/m³. The high DRF(2-hr) shown by curve R-20 of Figure 5 is a result of longer

agglomeration time which produced a larger MMD aerosol by the time the blower was started.

Blower induced turbulence appeared to cause agglomeration to proceed rapidly. Large visible particles, of the order of 100 μm or more, were seen uniformly deposited on horizontal surfaces near chamber view ports. It is difficult to determine whether most of the agglomeration occurs in the strong velocity gradients near the fan wheel or whether induced turbulence in other parts of the chamber causes most of the agglomeration.

Two tests of enhanced turbulent agglomeration were performed under conditions different from the series described above. In test R-38, Figure 6, 3 lbs. of sodium were burned and the rate of oxidation was increased by blowing air over the molten sodium. This resulted in a peak aerosol concentration of 12 gm/m^3 40 minutes after initiation of the fire. The 3,000 cfm recirculating blower with shroud in place was started at $t=43$ minutes when the aerosol concentration was still near 12 gm/m^3 . An abrupt concentration fall-off yielded a DRF(2-hr) of 56. This experiment, as did other turbulence tests at high peak mass concentrations, showed a trend toward higher DRF's with increasing peak mass concentrations (see Table 1). Theoretical analyses of turbulent agglomeration(8) predict this behavior. It may be concluded that turbulent agglomeration methods will be especially useful for treating accidents which produce high concentrations of sodium aerosols. Although less effective for lower aerosol concentrations, the process is self moderating, i.e., the effect is proportional to the need for aerosol reduction. When turbulent agglomeration test designated R-34 was performed with the blower shroud exhaust opening directed downward against an impaction surface located several inches from the outlet, a DRF(2-hr) of 7.6 was achieved. This value is in the range of DRF(2-hr)'s obtained with the shroud outlet pointed up or with the shroud removed. Inasmuch as only a small amount of deposited sodium was observed near the blower outlet, it has been judged to be a further piece of evidence that most of the cleaning effect in these tests results from enhanced sedimentation by turbulent agglomeration rather than by centrifugal air cleaning in the blower or impaction of particles on the floor or other chamber surfaces.

These turbulence tests demonstrate the DRF(2-hr) values in excess of 10 are readily achievable for vessels 4 meters in height. When these results are scaled to containment vessel dimensions, they yield DRF(2-hr)'s of the same magnitude provided the same level of turbulence can be induced throughout the larger volumes. This assumes that the turbulent agglomeration process will produce substantial numbers of particles in excess of 100 μm in a matter of minutes, a process which we believe has been observed in our tests. It is likely that these large particles produce a further benefit by scavenging smaller particles as they fall through the aerosol cloud. The lower limit DRF(2-hr) for scale-up to a 30 m chamber can be found by imagining filling the 30 m vessel with a monodisperse unity density aerosol of diameter sufficient to produce the measured DRF(2-hr) in the 4 m high chamber. For example a DRF(2-hr) of 43 in the small chamber would scale to a DRF(2-hr) of 5 in a 30 m vessel.

POWDER DISPERSAL SCAVENGING TESTS

1. Limestone Powder and Sand Tests

To test the scavenging capacity of inert powders for a sodium aerosol we constructed a centrally mounted overhead dust spreader in our 90 m³ test chamber. The dust dispersal device consisted of a horizontal 16" diameter plastic disc rotated at 2,000 rpm by an electric motor mounted below it. Powder was dropped through a hole in the chamber ceiling onto the center of the disc and spread by rotational action throughout the chamber. An Acrison Model 120 dust feeder was used to feed powder to the rotating disc.

Initial tests employed crushed limestone (intended for agricultural use) which has a very low sodium content (< 1 ppm). The low sodium content of this powder permitted measurement of sodium aerosol particles on sampling filters by atomic absorption spectrophotometry without chemical interference from the scavenger dust. We recognized also that carbonates are widely used for suppression of sodium fires. During powder dispersal into a sodium aerosol cloud, the total mass of dust on the sampling filters rises considerably above the expected baseline level but the total sodium in the samples steadily declines as long as powder is injected.

Test R-25 and R-26, shown in Figures 7 and 8 demonstrate the cleaning effect of limestone powders when applied to the sodium aerosol in the manner described. The crushed limestone used in both tests had a MMD of 200 μ m and a GSD of 1.5. The total amount of dust, limestone plus sodium aerosol, rises above the normal baseline concentration of sodium alone during spreading. Not all the limestone particles are represented in the total mass concentration because of very inefficient capture of these large scavenging limestone particles by the sampling system. The concentrations for sodium aerosols alone, shown in Figures 7 and 8, were obtained by normalizing spectrophotometrically measured sodium mass collected on each filter with total aerosol mass concentrations from baseline initial samples.

In test R-25, powder was dispersed at a feed rate of 1.5 kg/minute for 21.5 minutes. The slope of the sodium concentration decay curve during this interval indicates a DRF(2-hr) of 6.0. In test R-26, powder was dispersed at the rate of 0.13 kg/min. for 36 minutes and was then boosted to 1.5 kg/min. for the next 60 minutes. The DRF(2-hr) associated with the slope of the different dispersal rate segments was 4.1 and 7.2, respectively. These data show that under similar dispersal conditions in a 30 m high containment vessel, identical, if not better, DRF(2-hr)'s would be achieved. This is because 200 μ m particles fall 30 meters in a matter of seconds. After they have fallen below the initial 4 m for which we have data, they would still retain their initial unit scavenging efficiency, perhaps enhanced by the accumulated sodium aerosol coating. However, a serious caution must be added in extrapolating to larger vessels. Mass subsidence effects get worse with increasing dispersed powder cloud volume, which would have a deleterious effect on overall scavenging efficiency.

Commercially available children's playbox sand was also used in one test of powder dispersal scavenging. During a one-pound sodium fire (R-32) in which a peak aerosol mass concentration of 1.54 gm/m^3 was reached, sand was dispersed by the Acrison feeder and rotary spreader at 1.6 kg/min . The mass median diameter (MMD) of the sand particles by sieve analysis was found to be $300 \text{ }\mu\text{m}$, whereas the MMD of crushed limestone was $200 \text{ }\mu\text{m}$. A DRF(2-hr) of only 2.3 was achieved in the R-32 sand dispersal test. This demonstrated that scavenging efficiency per unit mass injected was smaller for the larger sand particles. A total of 150 lbs. of sand was dispersed in the latter test.

After a new auger and surrounding feed tube was purchased for the Acrison 120 dust feeder to extend the maximum dispersal rate from 1.5 kg/min . to 15 kg/min ., test R-39, shown in Figure 9, was conducted with a feed rate of 3 kg/min . (double the previous limestone dispersal rate) and a significantly higher DRF(2-hr) of 10.2 was observed. Still higher dispersion rates would be expected to show greater two hour dose reduction factors, until counter-productive mass subsidence effects of bulk cloud settling began to have major effects - decreasing relative motion between scavengers and aerosol particles.

Table 2 summarizes experimental conditions and data from the successful scavenging tests. From the calculated two-hour dose reduction factors it was possible to determine an "effective" particle scavenging efficiency for each test. The simple model on which the effective efficiency calculation is based assumes that scavenging particles are spread uniformly across the chamber. The model also assumes monodisperse scavenging particles of size equal to the sieved particle mass median diameter (MMD). Neither of these assumptions was valid in practice. Therefore, values above and below the theoretical individual particle scavenging efficiency could occur. In this manner, effective scavenging efficiency could even exceed 1.0, as was the case during part of test R-26 when limestone dispersion was only 0.13 kg/min . Mass subsidence effects due to high concentrations of scavengers, uneven spreading by the rotor, and clumping of limestone (incomplete dispersion) are responsible for non-ideal effects.

2. Calculation of Effective Scavenging Efficiency

Effective scavenging efficiency, ϵ , is calculated from a simple exponential attenuation model.

$$\frac{C(t)}{C_0} = \exp\left[-\frac{3}{2} \frac{M}{\rho_s} \frac{\epsilon h}{D_s \sqrt{V}} t\right] = e^{-\lambda t} \quad (2)$$

The decay constant, λ , for exponential decline is implied by the experimentally determined DRF(2-hr):

$$\text{DRF}(2\text{-hr}) = \frac{\lambda t}{1 - e^{-\lambda t}} \quad \text{for } t=2 \text{ hours} \quad (3)$$

The experimentally found DRF(2-hr) determines λ in Equation (3). Use of Equation (2) then gives the effective efficiency as

$$\epsilon = \frac{-2}{3} \frac{\rho_s}{M} \frac{D_s V}{h} \lambda \quad (4)$$

where ρ_s = handbook density of scavenging particles

M = mass injection rate of scavenging particles

D_s = MMD of scavenging particles

V = test chamber volume

h = chamber height

When the feed rate of limestone was 1.5 kg/min. in test R-25 and R-26 the measured effective efficiencies were 0.30 and 0.36, respectively. If the sodium aerosol particles were 3 to 4 μm , as our previous measurements indicated, the Langmuir theoretical collection efficiency(12) for spherical 200 μm scavenger particles lies between 0.2 and 0.35. The lower limestone injection rate test (0.13 kg/min.) yielded an effective collection efficiency of 2.3. This efficiency was calculated by assuming the scavenger particle diameter to be 200 μm . The fact that the efficiency is greater than 1.0 seems to indicate that much of the aerosol collection was accomplished by the scavenger fraction less than 200 μm . For example, the theoretical collection efficiency of a 100 μm scavenger particle for 3-4 μm aerosol particles is still a fairly substantial 0.15 to 0.30. Perhaps the decreased mass subsidence effects at the lower dispersal rate permits much more efficient utilization of the broad range of scavengers.

Test R-39 at the much higher limestone dispersal rate of 3 kg/min., achieved a DRF(2-hr) of 10.2, yet showed an effective particle scavenging efficiency, 0.25, measurably less than for the lower dispersal rate tests. It may be concluded that higher DRF's are achievable with powder dispersal scavenging, but that decreases in per unit mass scavenging efficiency with higher dispersal rates will require large powder storage facilities. Perhaps more uniform areal dispersion mechanisms than were employed in the present tests would enhance unit mass scavenging efficiency at high dispersion rates.

The theoretical collection efficiency of 300 μm sand particles would normally be between 0.2 and 0.4. The experimentally determined value of 0.11 seems to result from non-uniform dispersal and more of the scavenging burden being shifted to particles smaller than 300 μm .

COMBINED POWDER DISPERSAL AND TURBULENCE TESTS

Several tests were conducted that combined the cleaning mechanisms of scavenging and turbulent agglomeration. In tests R-29 and R-31, 1 lb. of sodium was burned under standard conditions and then turbulence and scavenging with crushed limestone (MMD = 200 μm) were applied; first, successively and then, simultaneously. In test R-29, the blower was turned on for 8 minutes after a peak concentration of 1.5 gm/m³ had been reached. Immediately after the blower was shut down, 200 lbs. of limestone powder was dispersed at 1.5 kg/min. The objective was to "grow" larger target aerosol particles

which would be more readily collected by the scavenging limestone particles. A DRF(2-hr) of only 5.1 was achieved, a smaller figure than to be expected from either turbulence or powder dispersal separately.

Test R-31, on the other hand, was highly successful. It showed that simultaneous turbulent agglomeration and power dispersal scavenging could lead to a DRF(2-hr) greater than could be attained with either mechanism separately. Figure 10 illustrates the decrease of suspended sodium concentration during test R-31 in which a DRF(2-hr) of 20 was attained. The blower was operated continuously from $t=31$ minutes, when the total aerosol concentration was about 2 gm/m^3 , and 200 lbs. of crushed limestone was dispersed at 1.5 kg/min .

The increased relative motion between scavenging particles and aerosol particles produced by the turbulence, along with better dispersion of scavenger dust because of turbulence, explains the improved DRF in this test. DRF(2-hr)'s for turbulent agglomeration and scavenging, performed separately under the same conditions, would each be about 8.

ACOUSTIC AGGLOMERATION TESTS

The potential advantages of acoustic agglomeration for a direct-acting emergency air cleaning system have drawn increasing attention.(9,10,11) Sonic Fields exceeding sound pressure levels of 140 dB are known to produce rapid coagulation effects on high concentration aerosols.(9) Most experimental data and theoretical treatments have dealt with standing sound waves in specially tuned columns. Direct application of sonic agglomeration to a large, irregularly shaped containment vessel atmosphere could not be achieved conveniently with standing waves. It is more appropriate to envision a high intensity reverberant acoustic field throughout the vessel. Such a field could be produced by high intensity sound sources such as sirens, pulse jets, or whistles. Non-standing wave fields, or progressive acoustic waves, are, from theoretical considerations, thought to "sweep" the aerosol particle size distribution and bring about orthokinetic motion and coagulation between a broad range of size classes.(10)

Efforts to determine the effect of acoustic agglomeration on sodium combustion aerosols began with the search for a suitable and convenient source of high intensity sound with a proven ability to agglomerate aerosols in a similar size regime. The first sound generating devices employed in this research were pulse jet engines of the type used by Scott and his co-workers at the University of Toronto.(10) Considerable experimental difficulties were encountered when attempting to use pulse jets to establish a high intensity acoustic field in the 90 m^3 chamber. Since no definitive acoustic agglomeration results were observed from these tests, discussion of the pulse jet experiments has been assigned to Appendix A.

To overcome the problems we experienced with pulse jets, we selected an electronic siren of the type employed in emergency vehicles to generate intense sound fields. A "Federal" brand model PA 20A amplifier and CJ 24 siren were used for tests conducted in a steel chamber 0.65 m^3 in volume with an average internal height of 75 cm (Figure 11). This smaller chamber was used, rather than the 90 m^3 test cell, because the acoustic

power radiated by a single CJ 24 siren is inadequate to produce high sound levels throughout the larger chamber. The CJ 24 "siren" is not a gas-dynamic rotating siren, as in the usual definition of a siren, but a very powerful loud speaker powered by an amplifier and signal generator. When 60 watts of electrical power were used to energize this siren, sound pressure levels of no less than 145 dB were measured throughout the 0.65 m³ chamber. The siren was used in the "yelp" signal mode, a complex wave form in the audible range which has a superimposed oscillation of approximately 5 Hz. To the shielded human ear, this sound appears to be nearly invariant in intensity.

In a typical test, between 50 and 100 gm of sodium metal were heated in a steel melt pot with an externally placed Bunsen burner at the base of the chamber. After the melted sodium started to burn in the sealed chamber, the entire volume was rapidly filled with a very dense sodium aerosol. Sodium evolution ceased after about 10 minutes. Open-faced glass filter samples were obtained at mid-chamber height to determine the mass concentration over time for both the unperturbed and siren-on conditions. 9.2 liter samples were taken (1.4% of the chamber volume) to minimize depletion of the aerosol by sampling. Because unperturbed and siren-on tests were sampled at roughly the same time intervals, changes in concentration due to sampling were identical for each.

Figure 12 shows mass concentration decay results for unperturbed sedimentation in the small chamber and sedimentation enhanced by siren induced acoustic agglomeration. The curves for the three sonic tests indicate marked enhancement of sedimentation.

The results of our acoustic agglomeration tests can be compared quantitatively in several ways. From initial mass concentrations between 8 and 21 gm/m³, unperturbed settling curves decay to 0.5 gm/m³ in about an hour. During the sonic tests, the decay curves reached the 0.5 gm/m³ level in only 20 to 30 minutes. Table 3 shows two-hour dose reduction factors computed from the data from this 75 cm high vessel. For similar peak concentration, the sonic tests show DRF(2-hr)'s from 2 to 3 times greater than the unperturbed tests. These tests were deliberately conducted at high initial mass concentrations in the 90 m³ vessel to increase chances of producing measurable sonic agglomeration.

Scale-up and comparisons with previous results in the larger chamber are difficult. It can be shown (Appendix B) that when comparing the stirred settling of the same monodisperse aerosol in chambers of different heights, the associated DRF(2-hr) in each case is approximately inversely proportional to height. (The only restriction is that when chamber height becomes sufficiently large that the DRF(2-hr) scaled-up by this method falls below about 2, large errors are introduced. The approximation made in Appendix B is no longer valid.) Although we did not have monodisperse aerosols in tests in the small chamber, (measured MMD was 3.9 μ m and GSD was 2.0), a fitted straight line to the semi-logarithmically plotted decay data yields an average particle sedimentation diameter. Therefore, an inverse - with-height scale-up nevertheless works reasonably well for a polydisperse aerosol. Scale-up of DRF(2-hr)'s for tests A-4, A-5, and A-6 in the 75 cm high chamber to the 4 meter high, 90 m³ chamber give DRF(2-hr)'s of 5.8, 3.1, and 5.0, respectively. Because peak mass concentrations were so much greater in the smaller settling chamber, these DRF's are in truth less impressive than might be assumed

at first glance. This is because high concentration aerosols have associated DRF(2-hr)'s for undisturbed settling which can be greater than the DRF(2-hr) of 1.4 found for baseline aerosols in the 90 m³ chamber. The single very high concentration undisturbed settling test conducted in the 90 m³ chamber (R-5) reached a peak concentration of 8.9 gm/m³ and had an associated DRF(2-hr) of 2.3. By this measure, acoustic tests A-4, A-5, and A-6 imply improved dose reduction performance when scaled-up, but not by the margin an initial impression would warrant.

CONCLUSIONS AND RECOMMENDATIONS

Three separate direct in-vessel air cleaning systems have been tested in prototype form over a range of test aerosol concentrations. Two of these systems, turbulent agglomeration and powder dispersal scavenging, have been tested concurrently and in tandem. The synergistic effect of these two systems, when operated concurrently, was found to be beneficial. Enhancing aerosol sedimentation by means of turbulent agglomeration was shown to yield greater dose reduction factors with higher aerosol concentrations - a result fully supported by aerosol agglomeration theory.

The powder dispersal scavenging system tested in a 4 m high chamber would show the same or better dose reduction performance in a 30 m containment vessel. A two-hour dose reduction factor exceeding 10 has been achieved. It is more difficult to scale-up the performance of turbulent agglomeration to a 30 m vessel without more thorough studies in vessels of different height and with varied levels of turbulence. Nevertheless it seems likely that if equivalent turbulence levels were established throughout a larger vessel, the dose reduction factor would not differ markedly from that found in the vessel of 90 m³.

Acoustic agglomeration of sodium combustion aerosols has been demonstrated. The experiments we conducted were unlike most sonic agglomeration tests reported in the literature in that progressive waves were set up in a large irregular volume rather than in a duct. Sirens were found to have far fewer operational difficulties than pulse jet engines as sources of high intensity sound for this application. Inasmuch as the initial sonic agglomeration tests were, of necessity, performed in a small vessel, and at high mass concentrations to compensate for sound levels not reaching the ideal 150-160 dB level, satisfactory comparison with other direct application methods is not possible at this time. It is significant that the feasibility of using acoustic agglomeration to induce rapid sedimentation of sodium aerosols has been proved in a small vessel.

Further experiments and development should focus on testing direct in-vessel methods, already shown feasible, i.e., turbulence, acoustic agglomeration, and powder dispersal, over a range of aerosol concentrations and at elevated temperatures. In general, these systems can be expected to function much better at higher initial aerosol concentrations. At low aerosol concentrations, the performance would be less efficient but, of course, at low aerosol concentrations less reductions are required to meet off-site standards. It is important to establish

quantitatively these concentration effects. The introduction of elevated temperatures would simulate accident conditions more realistically.

Turbulent agglomeration needs to be demonstrated at a variety of turbulence levels so that energy requirements for this approach can be minimized for vessels of containment size. A wider range of powder dispersal rates combined with improved dispersal methods should be examined. At the same time, more uniform areal deposition of the powder should be provided by using multiple dispersal devices. Acoustic agglomeration tests using sirens should be conducted in large vessels, and sound pressure levels should be raised 10 to 20 dB above the 145 dB level used in this study. Duration of sound exposure is an important parameter for future studies.

Other potential direct in-vessel cleaning methods, not tested in this study, include electrostatic precipitation, ESP combined with induced turbulence, and other methods of inducing turbulence, e.g., cyclones and vertex generators. These approaches also merit investigation.

TABLE 1

Effectiveness of Turbulent Agglomeration Related to
Sodium Aerosol Peak Mass Concentration

<u>Test #'s</u>	<u>Peak Mass Concentration (gm/m³)</u>	<u>DRF(2-hr)*</u>
14,15,16, 18,19,34	1.2 - 2.3	4.2 - 8.2
20,21,22	3.6 - 4.6	25 - 40
38	12	56

*Two hour dose reduction factor

TABLE 2

Powder Dispersal Test Conditions and Scavenging Efficiency

<u>Test #</u>	<u>Scavenging Particles</u>	<u>Dispersal Rate</u>	<u>DRF(2-hr)*</u>	<u>Effective Scavenging Efficiency**</u>
R-25	Limestone, 200 μ m MMD	1.5 kg/min.	6.0	0.30
R-26A	Limestone, 200 μ m MMD	0.13 kg/min.	4.1	2.3
R-26B	Limestone, 200 μ m MMD	1.5 kg/min.	7.2	0.36
R-32	Sand, 300 μ m MMD	1.6 kg/min	2.3	0.11
R-39	Limestone, 200 μ m MMD	3.0 kg/min.	10.2	0.25

*Two hour dose reduction factor

**See text

TABLE 3

Two-Hour Dose Reduction Factors in 0.65m³ Settling
Chamber for Acoustic Agglomeration and Baseline Tests

<u>Test #</u>	<u>Peak Aerosol Mass Concentration (gm/cm³)</u>	<u>DRF(2 hr)</u>
<u>Unperturbed Settling Tests</u>		
A-1	8.2	5.1
A-2	21.5	9.8
A-3	16.1	8.6
<u>Siren-On Tests</u>		
A-4	16.2	31.1
A-5	10.5	16.6
A-6	16.1	26.5

APPENDIX A

Tests of Pulse Jet Engines for Acoustic Agglomeration

Following the novel experimental approach to agglomerating zinc oxide fume used by David Scott(11) at the University of Toronto in Canada, we first chose to examine pulse jet engines as efficient high intensity sound generators. Scott and his co-workers demonstrated that exposing a ZnO aerosol with a MMD of approximately $1.5\text{ }\mu\text{m}$ to 160 to 165 dB sound from a 200 to 300 Hz pulse jet engine for several seconds resulted in an upward shift of the aerosol MMD to between $4.5\text{ }\mu\text{m}$ and $6\text{ }\mu\text{m}$. It is important to note that these results were related to tests with the pulse jet engine exhaust stream directed into a narrow circular duct through which the ZnO aerosol flowed. Upstream and downstream measurements of the aerosol size distribution gave the noted results for this continuous flow system.

It was not possible to dump the very hot and voluminous exhaust stream of the jet into the 90 m^3 HACL chamber because over heating, chamber pressurization, a need to vent excess pressure, and the presence of stray turbulent eddies from the exhaust stream would affect the aerosol. A special test geometry was required to irradiate the test chamber volume with high intensity sound without simultaneously disturbing the aerosol with jet exhaust and making it impossible to resolve the effects of sonic agglomeration alone. A Dyna-Jet "Red-Head"* pulse jet engine was tested in a U-shaped duct commuting with the atmosphere external to the 90 m^3 chamber. The sound transmission element was an 8" diameter 2 mil thick stainless steel duct segment which allowed the 240 Hz finite amplitude wave train to penetrate the foil membrane with less than a 10 dB sound pressure level reduction. Sound levels up to 138 dB near the transmission membrane were measured, but operation of the jet for longer than several minutes led to valve and combustion tube failure. No tests were conducted with this jet with sodium aerosol in the 90 m^3 chamber.

A more reliable pulse jet engine was acquired from the metallurgy section of the Ontario Research Foundation (ORF) near Toronto, Canada. We were unable to achieve sound levels over 135 dB more than 1 ft from the jet axis near the exhaust. A large thin film plastic tent (10'x6'x5') was set up in the 90 m^3 chamber so that, operated within, the ORF jet acoustic power would suffer minimum reduction transmission. Cooling air was provided via ductwork to and from the tent.

The mass concentration decay during test R-42 in which the ORF jet was operated continuously for one hour is shown in Figure 13. No significant departure was observed from the unperturbed decay measured in test R-45, also shown in Figure 13. From this comparison, we conclude that acoustic agglomeration under these conditions was negligible and that even extended exposure of the aerosol to 118 dB to 126 dB level sound in a reverberant chamber would not enhance sedimentation at this concentration level. The use of pulse jets to generate high intensity acoustic fields throughout a large volume appears to be impractical due to a multitude of operational difficulties, unless the jet exhaust can be directed into the vessel. This seems to be impractical for the application under consideration as it would increase pressurization of the containment vessel.

APPENDIX B

Scaling DRF's for Chambers of Varying Height for Stirred
Settling of Monodisperse Aerosols

The associated two-hour dose reduction factor for a monodisperse aerosol undergoing stirred settling in a chamber of height, h , is:(1)

$$\text{DRF} = \frac{\frac{K t_*}{h}}{1 - e^{-\frac{K t_*}{h}}} \quad (t_* = 2 \text{ hr}) \quad (\text{B-1})$$

$$\text{where } K = \frac{\rho d^2 g C_c}{18\eta}$$

d = aerosol particle diameter

ρ = aerosol density

g = gravitational acceleration

C_c = slip correction

η = gas viscosity

For $\frac{K t_*}{h} \gg 1.0$,

$$\text{DRF} \approx \frac{K t_*}{h} \quad (\text{B-2})$$

Therefore DRF and chamber height, h , are inversely proportional. For $\frac{K t_*}{h} = 2.0$ a 16% error in DRF is incurred by approximating the DRF as in B-2. When $\frac{K t_*}{h} = 1$, a 58% error is incurred.

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FIGURE 1

BASELINE CONCENTRATION DECAY PROFILE FOR 1.0 LB.
SODIUM FIRES IN 90 M³ AIR-FILLED CHAMBER

FIGURE 2

EIGHT-STAGE ANDERSEN IMPACTOR RESULTS FOR A 1.0 LB. BASELINE SODIUM FIRE

MEASURED TOTAL AEROSOL MASS CONCENTRATION/SODIUM MASS CONC.

FIGURE 3.

RATIO OF TOTAL AEROSOL MASS TO SODIUM MASS AS A FUNCTION OF TIME

FIGURE 4.

TURBULENT AGGLOMERATION AEROSOL CLEARANCE TESTS (LOW CONC.)

FIGURE 5.

TURBULENT AGGLOMERATION TESTS AT HIGH AEROSOL CONCENTRATION

FIGURE 6.TURBULENT AGGLOMERATION TEST AT 12 GM/M^3 AEROSOL CONCENTRATION

FIGURE 7

CRUSHED LIMESTONE POWDER DISPERSAL TEST NUMBER R-25

FIGURE 8.

CRUSHED LIMESTONE POWDER DISPERSAL TEST NUMBER R-26

29.
FIGURE 9.

30.
FIGURE 10.

TEST COMBINING TURBULENCE AND LIMESTONE POWDER DISPERSAL

FIGURE 11.

0.65M³ CHAMBER FOR ACOUSTIC AGGLOMERATION EXPERIMENTS WITH A SIREN

FIGURE 12.

ACOUSTIC AGGLOMERATION TESTS IN 0.65 M³ CHAMBER.

FIGURE 13.

ACOUSTIC AGGLOMERATION TEST IN 90 M³ CHAMBER WITH ORF PULSE JET