
Compilation of Contract Research for the Chemical Engineering Branch, Division of Engineering Technology

Annual Report for FY 1985

**U.S. Nuclear Regulatory
Commission**

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Compilation of Contract Research for the Chemical Engineering Branch, Division of Engineering Technology

Annual Report for FY 1985

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Compilation of Contract Research
for the
Chemical Engineering Branch
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Introduction

This compilation of annual research reports by the contractors to the Chemical Engineering Branch, DET, is published to disseminate information from ongoing programs and covers research conducted during Fiscal Year 1985. The programs covered in this document include research on: 1) engineered safety feature (ESF) system effectiveness in terms of fission product retention under severe accident conditions; 2) effectiveness and safety aspects of selected decontamination methods; 3) decontamination impacts on solidification and waste disposal; 4) evaluation of nuclear facility decommissioning projects and concepts, and 5) operational schemes to prevent or mitigate the effects of hydrogen combustion during LWR accidents.

The overall objectives of each of the programs reported on in this compilation are noted below:

1) ESF System Effectiveness - Battelle Pacific Northwest Laboratory

In evaluating potential releases of radioactivity from containment atmospheres to the environment as a result of severe light water reactor accidents, the effectiveness of certain Engineered Safety Features for removal of radioactive particulates is an important factor. This project will develop and validate codes and models to estimate particulate removal effectiveness.

2) Effectiveness and Safety of Decontamination Methods -
Idaho National Engineering Laboratory

The increased utilization of in situ decontamination of primary coolant systems and associated equipment has raised questions within the Nuclear Regulatory Commission as to the effectiveness of the decontaminations in reducing occupational exposure. This program is to obtain information, by collecting actual operating experience, on decontamination methods in reducing occupational exposures, waste forms generated, and potential problem areas.

3) Decontamination Impacts Solid and Waste Disposal -
Brookhaven National Laboratory

There has been an increase in the use of in situ decontamination of primary coolant systems which has raised questions regarding disposal of wastes from these decontaminations. This program is to provide a rational basis for licensing decisions concerning the impact of chemical solutions used for LWR

decontamination on waste solidification and disposal. This basis is to be developed through an evaluation of potential problems caused by these decontamination solutions, in particular chelating agents, and chemical processes which can be used to alleviate problems.

4) Facilities Undergoing Decommissioning -
United Nuclear Corp.

This project provides information to determine the proper amount of funds which must be available to ensure timely and safe decommissioning operations; also it provides information that can be used for confirmatory purposes in critiquing licensee decommissioning plans. This information is to be used by NRR for evaluation of reactor decommissioning plans and will be used by RES in the development of standards.

5) Hydrogen Control - Sandia National Laboratory

An igniter system is employed in a reactor containment to control the combustion of hydrogen that may result from certain possible accidents. Data are needed to reduce the uncertainties associated with the capability of an igniter system to perform its function, i.e., controlled burning of hydrogen without detonation, in the presence of water sprays provided to reduce the temperature of the containment atmosphere. Too much water on the igniter element would prevent the hydrogen-air mixture from reaching its ignition temperature. This project will provide the NRC licensing staff the information to evaluate the igniter systems and operational schemes proposed by reactor licensees.

ANNUAL REPORT

FISSION PRODUCT RETENTION EFFECTIVENESS
OF ENGINEERED SAFETY FEATURE SYSTEMS
DURING SEVERE ACCIDENTS

FIN B2444: Effectiveness of LWR
ESF Systems Under Severe Accident
Conditions

FIN B2884: ICEDF Code Verification/
Validation Tests

Work Supported by the
U.S. Nuclear Regulatory Commission
under Contract DE-AC06-76RLO 1830
Fin: B2444 and B2884

Pacific Northwest Laboratory
Richland, Washington 99352

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EFFECTIVENESS OF LWR ESF SYSTEMS UNDER SEVERE ACCIDENT CONDITIONS

PRINCIPAL INVESTIGATORS: W. K. Winegardner and P. C. Owczarski

FIN B2444

OBJECTIVE

The objective of this research is to evaluate light-water reactor (LWR) engineered safety feature (ESF) system effectiveness in terms of fission product retention to define the level of protection currently afforded and to determine how this level can be increased. System performance during postulated severe core damage/melt accident sequences is emphasized. Development of verified/validated analytical models to predict the effectiveness of spray, suppression pool, ice condenser, containment cooler, and containment and secondary auxiliary filtration systems for accident consequence analyses is a major part of the objective. The identification of any experimental investigations needed to validate proposed models and identify failure modes and effects is included. Also included are efforts to provide guidelines for system design and operating requirements, to identify possible add-on features, to identify the more important systems (and provide information for their emphasis in future regulatory processes), and to identify information gaps.

FY 1985 SCOPE

FY 1985 efforts focused on continued participation in U.S. Nuclear Regulatory Commission (NRC) staff and contractor efforts to reassess and update analytical procedures for estimating accident source terms for nuclear power plants. Specifically, work continued on the development of two computer codes to estimate the extent of particle retention in boiling-water reactor (BWR) pressure suppression pools and the ice compartments of pressurized-water reactor (PWR) ice condenser containment systems. The two codes, SPARC and ICEDF, respectively, have been included in the suite of computer codes that is being used to calculate source terms for selected plants and accident sequences (Silberberg et al. 1985). Data were made available that permitted interim SPARC code validation efforts. Development of a computer code (KOILDF) to estimate aerosol removal rates by containment recirculating air cooling systems continued. FY 1985 activities also included a review of the literature concerning source terms in relation to air cleaning systems and air filtration system performance during the combined challenges of severe accidents.

SUMMARY OF RESEARCH PROGRESS

In this section, progress is reported on the development and validation of the SPARC computer code for use with pressure suppression pools; the development of the ICEDF code for use with ice compartments of ice condenser containment systems; the literature review concerning air filtration system performance; and the development of the KOILDF code for use with containment cooling systems.

Suppression Pool Model

Work continued on the development of the computer code SPARC for use in predicting the extent of particle retention in BWR pressure suppression pools. Validation efforts were included. Problems encountered as a result of the incorporation of a complex condensational particle growth model into the code were resolved. This addition, made during the previous year, was included because expansion work associated with rising bubbles can result in the cooling of bubble atmospheres, supersaturation, particle growth, and possible enhanced particle collection. Sensitivity studies were undertaken to investigate the effect of this growth phenomenon. Plots of pool decontamination factor for various values of inlet particle size and concentration are shown in Figures 1 and 2. The following conditions were assumed in the development of the two figures: 100°C inlet gas; 50 mole% air and steam; 12-ft pool depth; 100°C pool; 1 atm pressure (at pool surface); negligible entrance effects; bubble equivalent sphere diameter and ellipticity ratio of 1.0 cm and 1.5, respectively; and a log-normal distribution for inlet particles ($\sigma_g = 2$). Figure 1 is a plot of the log of the decontamination factor (DF_i , ratio of mass rate of i size particles entering pool to mass rate of i size particles leaving) as a function of the log of the inlet particle size (d_i , dry particle diameter) for various values of inlet particle concentration. Figure 2 summarizes the effects of concentration and size on retention in terms of an overall decontamination factor (DF_0 , ratio of mass rate of all particles entering pool to mass rate of all particles leaving). The increase in retention associated with the lower concentrations, especially for the larger diameters, is the result of

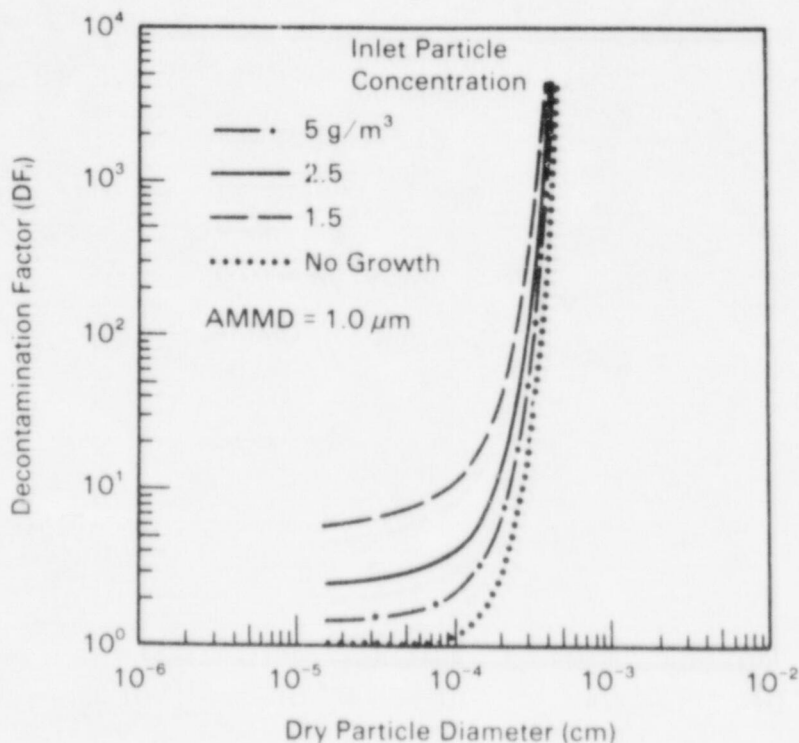


FIGURE 1. Pool Decontamination Factors Versus Particle Size

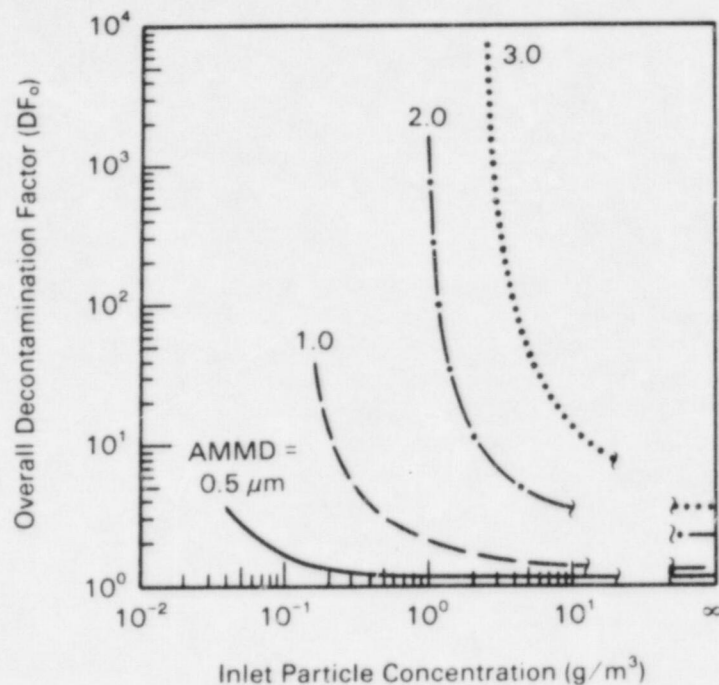


FIGURE 2. Pool Overall Decontamination Factors vs. Inlet Concentration

decreased competition between particles for the limited amount of supersaturated vapor that is available. In general, retention enhancement as a result of particle growth is predicted for concentrations below about 0.2 g/m^3 and for diameters greater than about $1 \text{ } \mu\text{m}$ aerodynamic mass median diameter (AMMD).

Other code modifications made in FY 1985 included the incorporation of improved correlations for representing the hydrodynamics of bubble swarms. Data were made available that were used in developing improved correlations for bubble size, shape, and swarm rise velocity. The correlation for bubble size was developed from swarm statistics and information reported by Paul et al. (1985, 1985a):

$$d_{vm} = 0.72 \exp \{2.303 [-0.2265 + (0.0203 + 0.0313 x_{nc})^{1/2}]\}$$

where d_{vm} is the bubble volume mean diameter and x_{nc} is the mole fraction of noncondensable gas entering the pool. For noncondensable gases only, the expression reduces to 0.72 cm . When steam accompanies the noncondensable gases entering the vent exit, the effective diameter decreases. A single value will be used to represent bubble size until validation efforts reveal that the use of actual swarm statistics is warranted. Data indicate that the swarms have an essentially log normal bubble size distribution that remains constant throughout the rise period because of the frequent coalescence and redispersion of the bubbles (for noncondensable gases only, the most frequent equivalent sphere diameter is 0.57 cm , with a standard deviation of 0.186).

Bubble shape is calculated using another correlation derived from the data of Paul et al. (1985, 1985a).

$$\frac{\text{major semiaxis (b)}}{\text{minor semiaxis (a)}} = 0.84107 + 1.13466 d_{vm} - 0.3795 d_{vm}^2$$

This correlation was established for $0.15 \text{ cm} \leq d_{vm} \leq 1.3 \text{ cm}$. Values of d_{vm} outside this range will give incorrect b/a values. All bubbles smaller than 0.15 cm are spheres ($b/a = 1.0$), while bubbles larger than 1.3 cm have $b/a \geq 1.67$ only in swarms. In fact, isolated, single, large bubbles can be much flatter and become lenticular when rising.

The residence time (t_b) of bubbles in the pool is determined by the bubble swarm rise velocity (V_{sw}) and vent depth (h_p). This residence time is an important parameter and is determined by

$$t_b = \int_0^{h_p} dx / V_{sw}$$

where x is the vertical coordinate from the vent exit to the pool surface. Swarm rise velocity is not constant along this coordinate. With the increase in the volumetric flow rate as the swarm rises, V_{sw} increases. V_{sw} represents the volumetric average velocity on a cross section of the swarm; bubbles in the center rise faster than swarm periphery bubbles. Using data from Paul et al. (1985a) and General Electric Company (1981), the following correlation was developed for swarm rise velocity:

$$V_{sw} = [Q_s + 5.33]/3.011 \text{ E-03}]^{1/2} (1 - 3.975 \text{ E-04 } x) \text{ (cm/s)}$$

where Q_s is the total gas volumetric flow rate (l/s) at depth $h_p/2$ (cm). An average swarm velocity is currently used in the SPARC code to simplify calculations:

$$\bar{V}_s = [V_s (h_p = 0) + V_s (h_p = \text{vent depth}/2)]$$

Initial model data comparisons were based on pool scrubbing experimental data reported by Cunnane et al. (1985) and the version of the code that included the preceding condensational particle growth models and the improved correlations for hydrodynamic processes. These comparisons resulted in calculated decontamination factor values that were consistently lower than those obtained by experiment (see Figure 3). Two additional particle capture mechanisms--deposition in bubbles from inertial forces initiated by vent exit gas velocities and bubble coalescence and redispersion--were proposed as a result of this poor correlation. Details of early models developed for the two retention processes, initially based on experimental constants, are discussed below.

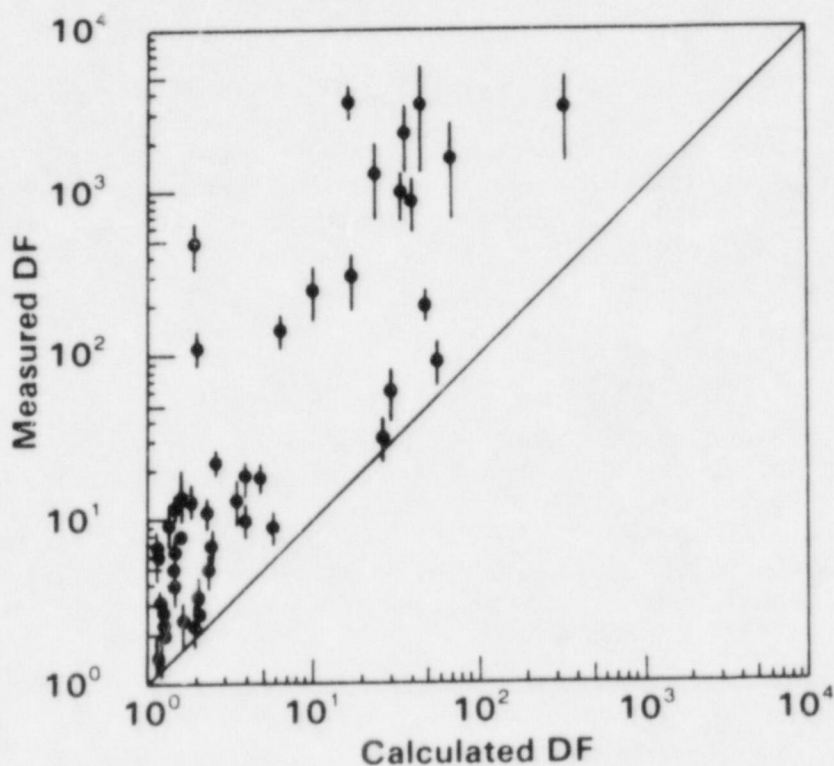


FIGURE 3. Model Data Comparison Prior to Addition of Inertial Deposition Models and Experimental Constant Optimization

The gas entering a forming bubble can circulate during initial bubble growth and release of the bubble to the pool. The horizontal injector used in the experiments resulted in considerable gas momentum in the forming globules, potentially resulting in particle scrubbing by inertial deposition from centrifugal forces. The forming globule scrubbing velocity for particle size i is a function of the injection rate. An analysis of globule volume and velocity suggests the following form for the expression for decontamination factor:

$$DF_i = \exp [A_1 V_s(i) V_e^{A_2}]$$

where A_1 and A_2 are experimental constants, $V_s(i)$ is the settling velocity of particle size i , and V_e is the vent exit velocity.

During swarm rise, redispersion or breakup could also cause bubble surface motion, centrifugal forces, and inertial deposition. The decontamination factor associated with this behavior was expressed using the following equation:

$$DF_i = \exp(E V_s(i) \gamma \Delta t / d_b g \mu)$$

where E is an experimental parameter related to the formation frequency of a bubble, γ is the liquid surface tension, μ is the liquid viscosity, d_b is the bubble equivalent volume sphere diameter, Δt is the total bubble rise time, and g is the acceleration of gravity. The parameter E depends on the swarm density, which, in turn, depends on the swarm volume flow rate, G_s . In SPARC, an empirical model assumes that

$$E = A_3 G_s^{A_4}$$

where A_3 and A_4 are experimental constants. Use of this equation for E results in a final expression for the decontamination factor that is similar to the one developed above for vent exit effects.

Incorporation of the above two particle removal models, followed by optimization of experimental constants, resulted in a noticeable improvement in agreement between calculated and experimental values (Figure 4). In fact,

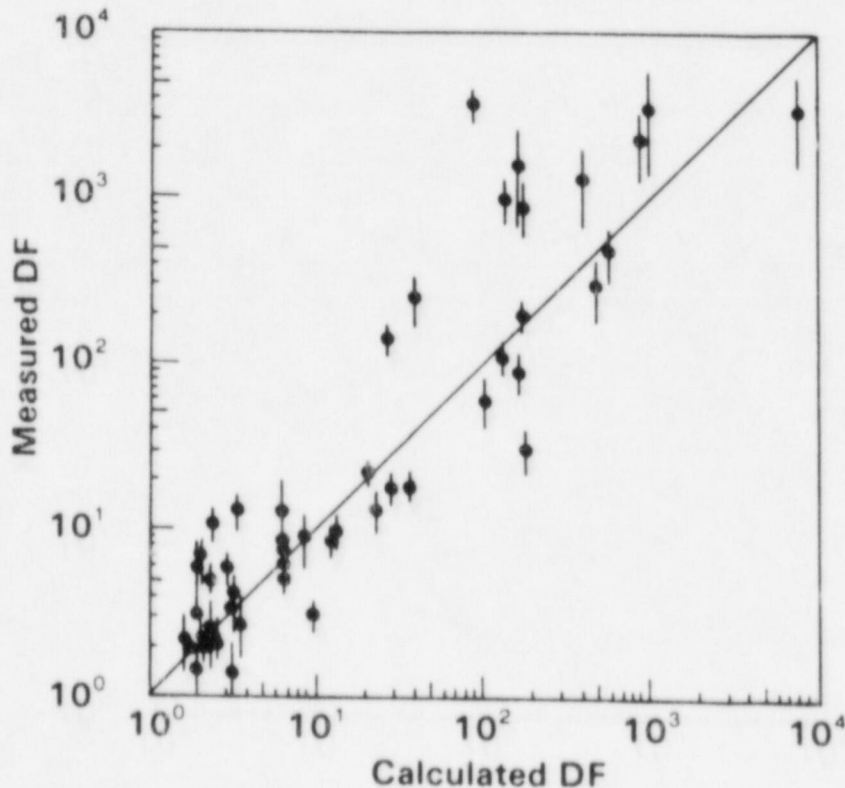


FIGURE 4. Model Data Comparison After Addition of Inertial Deposition Models and Experimental Constant Optimization

sensitivity studies revealed these as the two most important models. This is shown in Table 1, which lists decontamination factor values calculated for selected combinations of the various particle retention models.

Ice Compartment Model

FY 1985 efforts focused on the publication of a report (Owczarski et al. 1985) that describes the technical bases and use of the ICEDF computer code. This code is being developed to estimate the extent of particle retention in the ice compartments of PWR ice condenser containment systems. The section of the report describing technical bases focuses on particle retention and growth models. Retention mechanisms modeled include gravitational settling, impaction and interception, Brownian diffusion, diffusiophoresis, and thermophoresis. Condensational particle growth models for both supersaturated and unsaturated environments are described. The former is based on the principles of cloud physics coupled with detailed mass and energy balances. The latter was included because, in the case of soluble particles, condensation can occur in unsaturated conditions. The report is also intended to serve as a user's manual. It includes the following: a description of the code with methods/algorithms of calculation and subroutines; code operating instructions with input requirements, output descriptions, parameter selection guidelines, and examples of code use; a parameter sensitivity study summary; and a complete code listing.

The ICEDF code was used throughout the year to provide insights concerning the design of the engineering-scale experimental facility planned for use in code validation efforts (see FIN No. B2884, Summary of Research Progress, Engineering/Unit Cell Tests). Sensitivity studies investigated the effect of pressure on aerosol deposition. Figure 5 is a plot of decontamination factor as a function of the fraction of ice remaining for various assumed combinations of one or two atmospheres of ice compartment pressure and soluble or insoluble particles. Review of Figure 5 reveals that pressure appears to affect the decontamination factor much less than either particle solubility or the amount of ice present. Such results are important because they indicate that validation experiments can be performed at atmosphere pressure. Therefore, costly

TABLE 1. Calculated Decontamination Factors for Various Combinations of SPARC Particle Retention Models

| Retention Models | DF(a) |
|---|-------|
| All models operating | 15.8 |
| Redispersion/breakup during bubble rise only | 8.3 |
| Vent exit circulation only | 7.2 |
| Diffusion, settling, and circulation during bubble rise | 4.8 |
| Diffusion and settling during bubble rise | 3.4 |
| Circulation during bubble rise only | 3.1 |

(a) Geometric mean calculated from n runs from

$$[\prod_{i=1}^n \text{DF}(\text{calculated})_i]^{1/n}$$

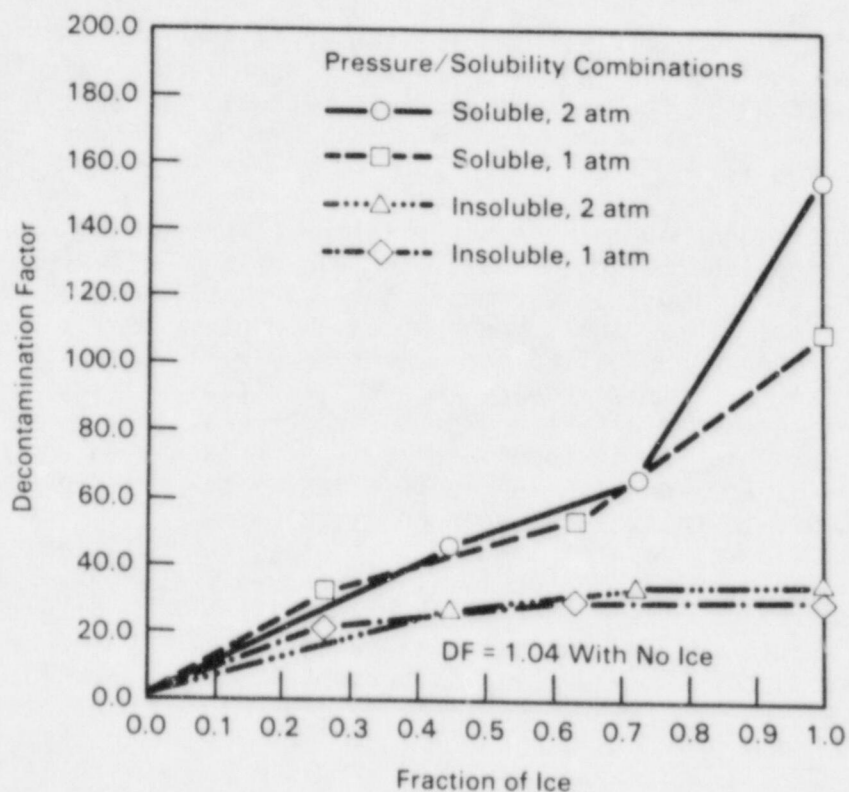


FIGURE 5. Decontamination Factor as a Function of Ice Remaining. Inlet conditions: temperature = 125°C; flow = 20 m³/s; steam mole fraction = 0.9; geometric mean particle diameter = 0.6 μm (σ_g = 2); particle density = 4.5 g/cm³; mass concentration = 2.0 g/m³.

pressurized facilities and aerosol sampling techniques will probably not be required. The code was also used to predict the extent of particle retention for matrices of candidate test conditions developed from statistical design efforts (test designs).

Filtration/Air Cleaning Systems

A review of the literature concerning source terms in relation to air cleaning systems and air filtration system performance during severe accidents was completed. Efforts focused on literature concerning radionuclide retention and/or release and system reliability and failure modes under adverse conditions. These conditions included elevated temperature and humidity and increased fission product loading and aerosol concentration.

The review revealed that air cleaning systems should be recognized as potentially significant source term mitigation systems, especially for secondary containment and auxiliary buildings. The severity of environmental conditions in these buildings can decrease to the point where significant particle retention and iodine adsorption may occur during all or portions of an

accident. On the other hand, the review also revealed that even if there are adequate predictions concerning the progression of the accident, there is insufficient information or test data to permit mechanistic modeling of system behavior under the combined challenges expected to occur in the event of a severe accident. However, some testing is being done to define the effects of combined challenges. The agenda of the recent (18th) DOE Nuclear Airborne Waste Management and Air Cleaning Conference included a session on High Efficiency Particle Air filter performance under adverse conditions, namely high heat and humidity (Bellamy et al. 1985).

Containment Cooling System Model

Development of the computer code KOILDF for use in estimating aerosol removal rates by containment recirculating air cooling systems continued. A draft report describing the technical bases and current status of the code was completed. Instructions for code operation were included. The code can be used for the analysis of both the heat and mass transport behavior of a cooling coil under conditions expected following a severe accident. It is equally applicable for calculations involving fin-tube coil or plate fin-and-tube coil designs. Four methods of aerosol scrubbing were considered in the development of the code: impaction, interception, thermophoretically augmented diffusion, and diffusiophoresis. The code can be used to estimate the transport behavior of an aerosol consisting of 20 discrete particle sizes. Temperature, moisture content, and aerosol composition are tracked on a macroscopic basis as the aerosol moves through the coil. The output of the program includes decontamination factors, penetration factors, and collection efficiencies for all 20 discrete particle sizes. Overall values for these parameters are also included.

FUTURE RESEARCH PLANS

As part of the continuing development of ESF system fission product retention models, plans for future research focus on computer code validation. Initial emphasis will be placed on the SPARC and ICEDF codes. In the case of the SPARC code, model data comparisons have been improved by the addition of equations derived from experimental observations. Plans include the development of a more mechanistic treatment. Additional pool scrubbing data should become available from tests using entrance geometries other than the one used in the initial series of tests. Such data should provide additional insights concerning the importance of hydrodynamic effects from gases entering the pool. Investigation of the potential for particle retention from bubble breakup and coalescence during the rise of dense swarms will continue. Underprediction of the extent of particle scrubbing in hot pools has been attributed to differences in circulation patterns. Improved representations of residence times for hot pools should also assist in the improvement of model data comparisons.

Experiments to provide validation data for the ICEDF code are scheduled to begin in FY 1987. Engineering-scale tests are planned with and without steam and/or ice to separate particle retention by gravitational and inertial effects from those associated with steam condensation.

Future plans involve examination of the need for new or improved models, including those used to describe fission product retention by spray systems. Performance in a condensing steam environment will be emphasized. Plans include expansion of the initial effort to model aerosol removal by containment recirculating air cooling systems to include consideration of experimental and field survey efforts that have been performed in attempts to characterize deposition and fouling on extended surface air cooling coils. Identification of the need for any new validation experiments is also planned. Although extensive parametric studies have not been performed, it is apparent that containment air coolers could be effective in removing aerosols from the containment air following a severe accident. However, the fate of the particles after they arrive at the collection surfaces is less certain. It cannot be easily determined if these particles would be washed off by the draining condensate or if they would accumulate and eventually plug the coil. Experiments to investigate the extent and rate of coil plugging should be planned. Planning of experiments to determine the effects of combined severe accident challenges on the performance of air cleaning/filtration systems is also needed.

To date, emphasis has been placed on fission products in the form of aerosols. It is also planned to incorporate absorption models for gaseous fission products into the ESF system retention computer codes. Finally, further investigation of the likelihood and extent of entrainment or conversion (to a volatile form) of previously captured fission products is planned.

ICEDF CODE VERIFICATION/VALIDATION TESTS

PRINCIPAL INVESTIGATORS: W. K. Winegardner and L. D. Kannberg
FIN B2884

OBJECTIVE

The objective of this research is to obtain experimental data that will aid in the development and validation of the ICEDF computer code. The code is part of the formulation of a systematic, mechanistic approach to predicting the extent of release of radionuclides from nuclear power plants under accident conditions (Gieseke et al. 1984). Specifically, the code is being used to predict the extent of particle retention in the ice compartments of PWR ice condenser containment systems during postulated core melt accident sequences. The development and improvement of calculational procedures used in making the above predictions is an evolutionary process; in the long term it must be verified through experimental studies. Much of aerosol retention theory has been validated by experiment. However, for the ice condenser system, the ice contributes major uncertainties and unvalidated phenomena. As a result, aerosol behavior in the presence of ice is being investigated. The effects of inlet gas composition, flow rate, temperature, and ice condition are all considered part of the objective of the project.

FY 1985 SCOPE

Initial FY 1985 activities focused on the definition of ICEDF code validation experimental methods, plans, and facilities and led to a two-phase approach that includes laboratory- or bench-scale investigations as well as engineering-scale tests. Subsequent to this definition, the scope was expanded to include the design, fabrication, and installation of the apparatus for the laboratory tests and the actual performance of tests. Activities associated with the engineering-scale tests included the identification of criteria, facility design, and the use of statistical methods to identify candidate test conditions.

SUMMARY OF RESEARCH PROGRESS

FY 1985 progress associated with the laboratory and engineering-scale ICEDF code validation experimental effort is detailed below.

Laboratory Tests

Progress included the completion of the design, fabrication, and installation of the laboratory test facility and the initiation of testing. Major purposes of the laboratory- or bench-scale tests are to refine and test experimental techniques and conduct investigations that will aid in the identification of key parameters for the engineering-scale investigations. The facility was designed to generate solid particles by nebulizing and drying solutions of

water-soluble materials or liquid suspensions of monodisperse latex spheres. Polydisperse aerosols can be further classified and sorted into essentially monodisperse distributions by techniques based on electrical mobility. Aerosols of the desired properties are then directed past a cylindrical deposition target. As shown in Figure 6, aerosol enters from the bottom of a chamber that contains the target via radially drilled holes, flows past the target, and is finally directed to instrumentation to detect particle concentration. Provisions have also been made so that steam can be introduced into the chamber along with the aerosol. An individual test consists of rapidly inserting the target into the chamber after the latter reaches steady state. Changes in effluent concentration associated with target insertion and removal are then observed.

In initial tests to determine the extent of background retention, no particle removal was observed upon inserting a room temperature target into the chamber. This early investigation was followed by an extensive test series to determine if retention could be enhanced by thermophoresis. Again, no particle capture was observed. Chilled targets as well as targets formed from ice were used in unsuccessful attempts to remove up to 10- μ m diameter particles from monodisperse, room temperature aerosols. Efforts to accelerate particle removal by heating aerosols to 100°C, thus increasing the temperature gradient across the target fluid boundary layer, were also unsuccessful.

Tests conducted at the end of the year confirm that particle removal by steam condensation will be a key factor in any large-scale tests. An obvious decrease in effluent concentration is observed when an ice target is inserted into steam-aerosol mixtures.

Engineering (Unit-Cell) Tests

FY 1985 research progress related to the engineering-scale experimental program included the development of design criteria, completion of a preliminary test facility design, and the development of candidate test designs (combinations of parameters for which tests are to be conducted). Details concerning these activities follow.

Design Criteria. Design criteria are intended to serve as targets for the development of test and test facility designs. The initial step in the development of the criteria was the determination of the conditions under which it is expected the ICEDF code will be applied. This step was accomplished by reviewing the results of the recent source term reassessment investigation for the PWR ice condenser containment design (Gieseke et al. 1984). Summaries of pertinent conditions predicted for the various postulated accident sequences associated with this investigation are shown in Tables 2 and 3 (lists of peak and time averages for significant inlet thermohydraulic and aerosol conditions, respectively). Review of the conditions predicted for the accident sequences coupled with design, cost, and schedule considerations and insights gained from ICEDF code calculations resulted in the criteria listed in Table 4. Comparison of the values listed in Table 4 with those presented in Tables 2 and 3 for the accident sequences indicates that, in most instances, the design criteria bracket the time average conditions. The most noticeable exception is pressure.

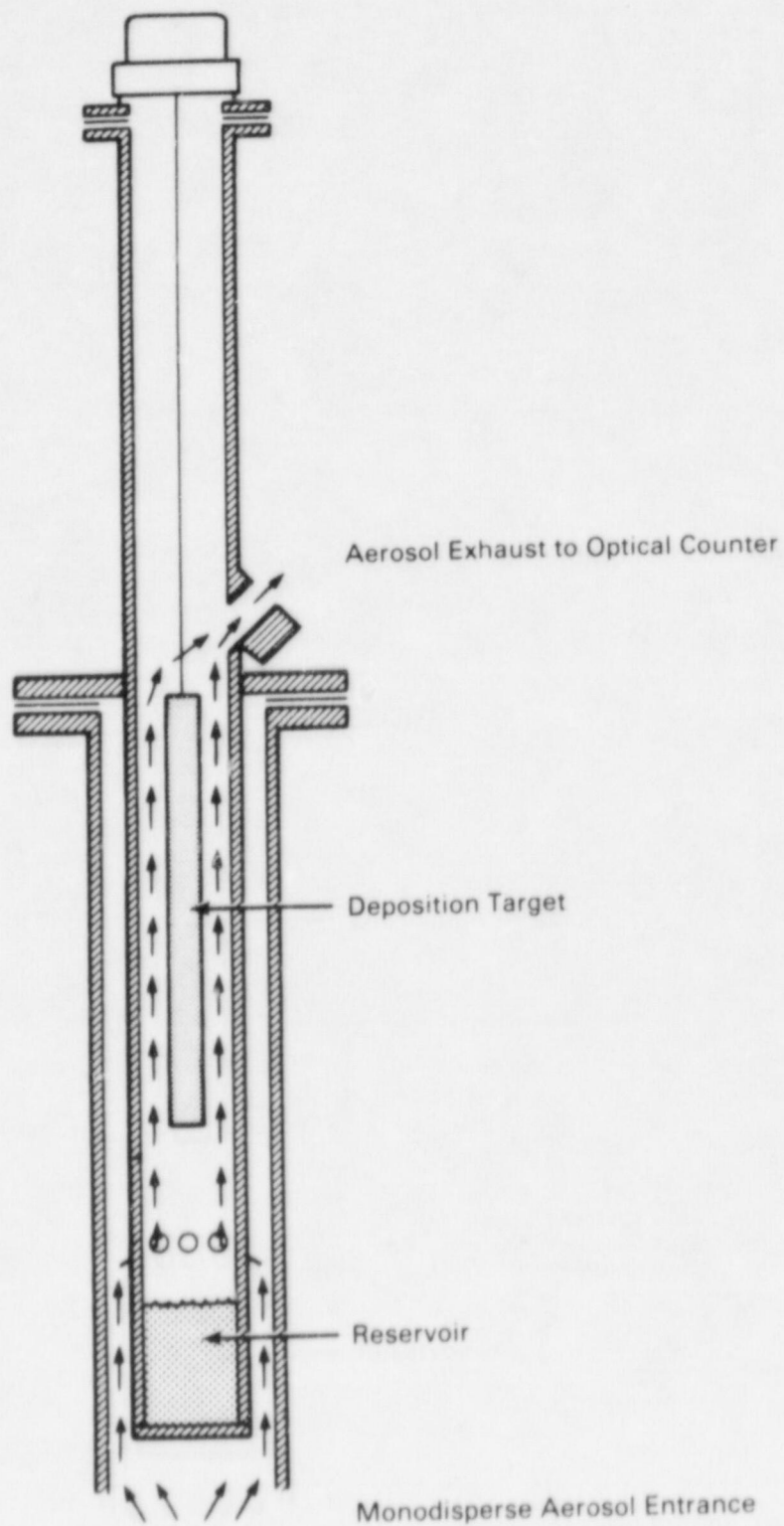


FIGURE 6. Ice Condenser Validation Tests, Phase 1 Laboratory Apparatus

TABLE 2. Thermohydraulic Conditions, PWR Ice Condenser Containment Design Accident Sequences

| Parameter | TMLB* | | TML | | S ₂ HF |
|---|---|--------------------|-----------------------|--------------------|--------------------|
| | Y | Δ | Y | Δ | |
| Inlet temperature, °C | | | | | |
| Time average ^(a) | 109 | 122 | 91 | 78 | 184 |
| Time average ^(b) | 122 | 130 | 91 | 77 | 112 |
| Maximum ^(c) | 1180 | 135 | 265 | 790 | 1430 |
| Minimum | 95 | 102 | 58 | 60 | 90 |
| Inlet gas flow rate, cm ³ /s | | | | | |
| Time average ^(a) | 1.58×10^7 | 2.51×10^7 | 2.55×10^8 | 9.28×10^6 | 5.34×10^6 |
| Maximum ^(c) | 2.13×10^9 | 1.8×10^9 | 1.55×10^{11} | 2.18×10^9 | 6.7×10^8 |
| Minimum | Periods of zero flow occur frequently in most scenarios | | | | |
| Inlet gas pressure, atm | | | | | |
| Time average ^(a) | 1.15 | 2.3 | 1.12 | 1.66 | 1.43 |
| Time average ^(b) | 1.29 | 2.9 | 1.12 | 1.41 | 1.33 |
| Maximum ^(c) | 5.7 | 4.1 | 4.5 | 3.5 | 8.1 |
| Minimum | 1.0 | 1.0 | 1.0 | 1.14 | 1.0 |
| Ice fraction | | | | | |
| Time average ^(a) | 0.16 | 0.2 | 0.33 | 0.17 | 0.003 |
| Time average ^(b) | 0.34 | 0.4 | 0.33 | 0.26 | 0.05 |
| Maximum ^(c) | 0.70 | 0.7 | 0.66 | 0.66 | 0.13 |
| Minimum | 0.0 | 0.0 | 0.08 | 0.14 | 0.0 *w* |
| Inlet water vapor | | | | | |
| Mole fraction ^(d) | | | | | |
| Time average ^(b) | 0.93 | 0.98 | 0.85 | 0.69 | 0.87 |
| Maximum ^(c) | 0.99 | 0.99 | 0.97 | 0.96 | 0.87 |
| Minimum | 0.73 | 0.92 | 0.54 | 0.14 | Unknown |

- (a) Time averaged over the entire simulation period.
 (b) Time averaged over only the period during which ice was in the ice condenser.
 (For TML Y, ice was present during the entire accident simulation).
 (c) Peak values are as computed with available codes. In some cases (such as gas flow for TML Y), computational artifacts are apparent due to the highly transient nature of reactor accident phenomena.
 (d) Water vapor mole fractions could be computed with the data available only for periods when ice was present in the ice condenser.

TABLE 3. Aerosol Conditions, PWR Ice Condenser Containment Designs Accident Sequences

| Parameter | TMLB ¹ | | TML | | S ₂ HF |
|--|---------------------|---------------------|------------------------|------------------------|---------------------|
| | Y | Δ | Y | Δ | |
| Time avg mass concentration, g/m ² | | | | | |
| Part time avg | 4.962 | 0.988 | 8.411×10^{-1} | 2.049 | 19.283 |
| Part time avg | 0.973 | 3.005 | 2.030×10^{-1} | 0.242 | 7.353 |
| Total time avg | 1.043 | 2.880 | 3.265×10^{-1} | 0.320 | 7.559 |
| Time avg aged count concentration, #/cm ² | | | | | |
| Part time avg | 2.836×10^5 | 7.036×10^4 | 1.837×10^5 | 1.138×10^5 | 5.126×10^5 |
| Part time avg | 2.509×10^6 | 2.567×10^6 | 2.681×10^6 | 2.323×10^6 | 2.387×10^6 |
| Total time avg | 2.470×10^6 | 2.459×10^6 | 2.197×10^6 | 2.228×10^6 | 2.355×10^6 |
| Time avg aged geometrical median diameter, μm | | | | | |
| Part time avg | 1.052 | 1.390 | 1.467 | 1.390 | 1.540 |
| Part time avg | 0.298 | 0.448 | 0.272 | 0.192 | 0.834 |
| Total time avg | 0.311 | 0.489 | 0.504 | 0.243 | 0.846 |
| Time avg release rate, g/s | | | | | |
| Part time avg | 2.099×10^2 | 8.41×10^1 | 8.41×10^1 | 8.41×10^1 | N/A |
| Part time avg | 8.350 | 8.35 | 2.841 | 9.226×10^{-1} | N/A |
| Total time avg | 1.191×10^1 | 1.142×10^1 | 1.858×10^1 | 4.494 | 4.316×10^1 |

Sensitivity tests with the ICEDF code indicate that pressure is one of the least important parameters (see FIN No. B2444, Summary of Research Progress, Ice Compartment Model).

Test Facility Design. Facility design is currently based on a full column of ice baskets, resulting in a 56-ft-high assembly. It is planned to install the facility in a building that has a 20-ft-diameter pit extending 50 ft below grade and an overhead crane height of 70 ft above grade. As shown in Figure 7 (an isometric view of the proposed facility), the necessary equipment for the test—including the air compressor, air heater, steam superheater, aerosol generator, aerosol/steam/air mixing chamber, bypass, inlet, and exhaust diffusion and turning vanes, and effluent scrubber—are located on various levels of the pit and on grade. A process flow diagram is shown in Figure 8.

The design of the facility test section is based on providing all the ice surface configurations and ice mass geometries that would be seen in a full-sized ice condenser, while minimizing the cross-sectional area. A full-sized ice condenser contains 1944 baskets, while the test section will contain an equivalent of four baskets. The cross section is shown in Figure 9; it is composed of one full basket, four half baskets, and four quarter baskets. A

TABLE 4. Thermohydraulic and Aerosol Variables and Their Upper and Lower Bounds

| Variable | Abbreviation | Lower Bound | Upper Bound |
|--|--------------|-----------------------|-----------------------|
| Thermohydraulic | | | |
| Inlet temperature, °C | T | 60 | 200 |
| Inlet gas flow rate, m ³ /s | FR | 15 | 200 |
| Gas pressure, atm | P | 1.0 | 1.0 |
| Ice fraction | IF | 1.0(a) | 1.0 |
| Inlet water vapor mole fraction | WF | 0.04 | 0.90 |
| Aerosol | | | |
| Particle solubility | PS | Soluble or Insoluble | |
| Count median diameter, μm | CMD | 0.08 | 2.0 |
| Geometric standard deviation, μm | GSD | 2.0 | 2.0 |
| Particle density, g/cm ³ | DEN | 1.0 | 4.5 |
| Mass concentration, g/m ³ | MC | 0.01 | 20.0 |
| Count concentration, #/cm ³ | CC | 5.0 x 10 ³ | 1.0 x 10 ⁶ |

- (a) For purposes of definition of the experimental region, the ice fraction is fixed at 1.0. However, the experiment will contain a single test case at which IF = 0 (no ice).

lattice framework is located every 6 ft to maintain vertical alignment and spacing of the baskets. The half and quarter baskets will be made by cutting full baskets, then welding a similar mesh to the back-side or sides. The mesh used for the baskets consists of 14-gage galvanized steel, with 1-in.-square holes on 1-1/4-in. centers. Basket sections are 12 ft high, and the total basket height is 48 ft. Internal cruciforms located adjacent to the lattice framework are used to stiffen the baskets. A screen and stiffening ring are added to the bottom of the ice basket. They not only provide support but also contain the ice and to permit air flow once the ice has melted. Approximately eleven 8-in. flanged windows will be installed for viewing events in the test section. The top halves of the windows contain a mechanical wiper motor and a light, and each window will provide a visual opening of 2 in. x 4 in. Flanged instrument probe ports will be located directly around the corner from the windows. Flanged access doors will also be located close to the windows. For the access doors to be useful, one half basket and possibly the full basket will be temporarily removed.

Pressurized air, steam, and other gases will be required during ICEDF model validation tests. The total flow rate of gas approaching the inlet of the test section will be constant for each test and will range from 0.029 to 0.38 m³/s over the complete set of ICEDF validation tests. This corresponds to prototypic flow rates ranging from 15 to 200 m³/s. Under most test conditions, the steam is expected to be removed from the flow by condensation. The result is that the flow rate above the condensation region will be reduced and is expected to range between 0.0029 and 0.38 m³/s.

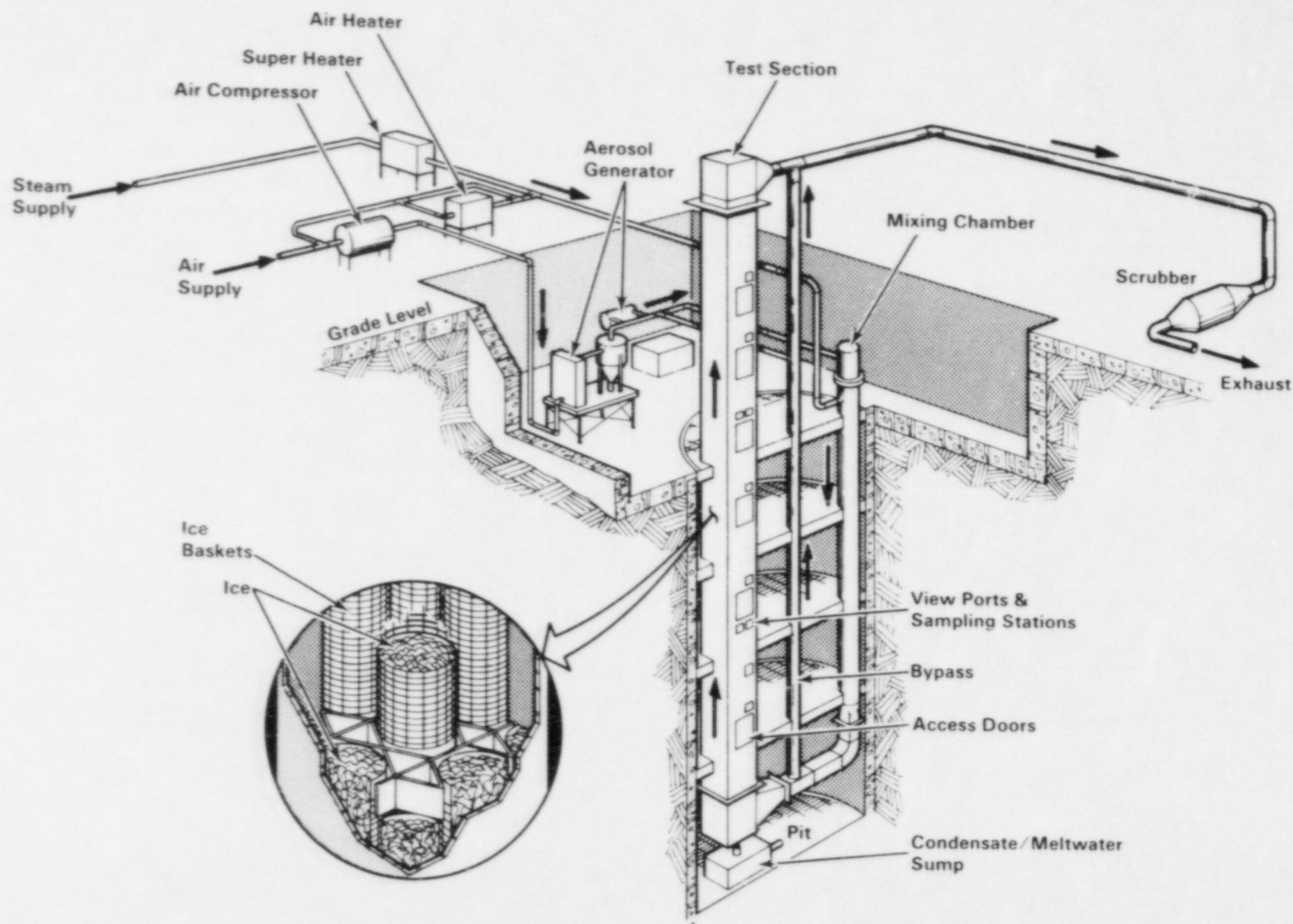


FIGURE 7. Isometric View of Proposed Aerosol Deposition Test Assembly

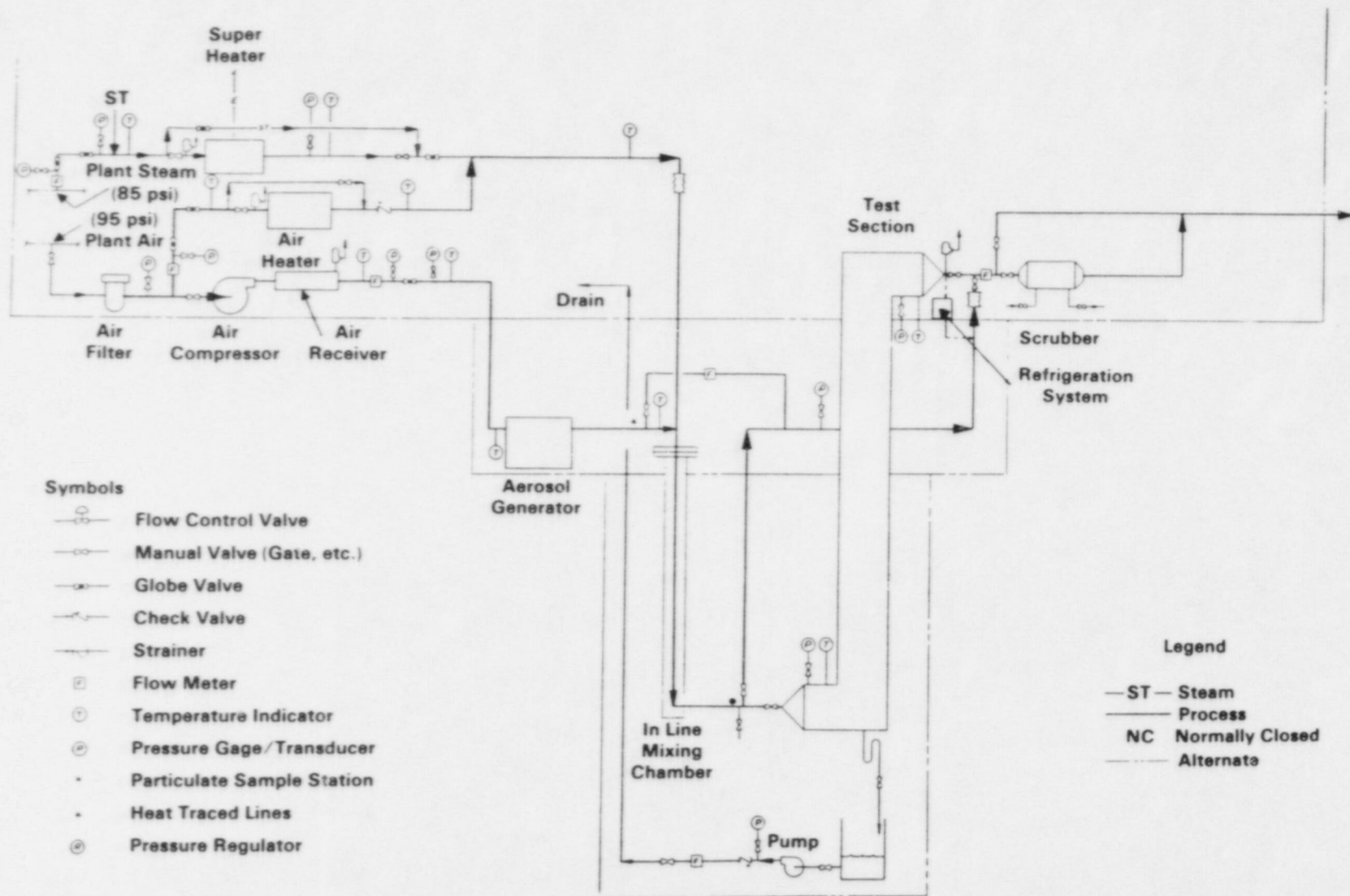


FIGURE 8. Process Flow Diagram

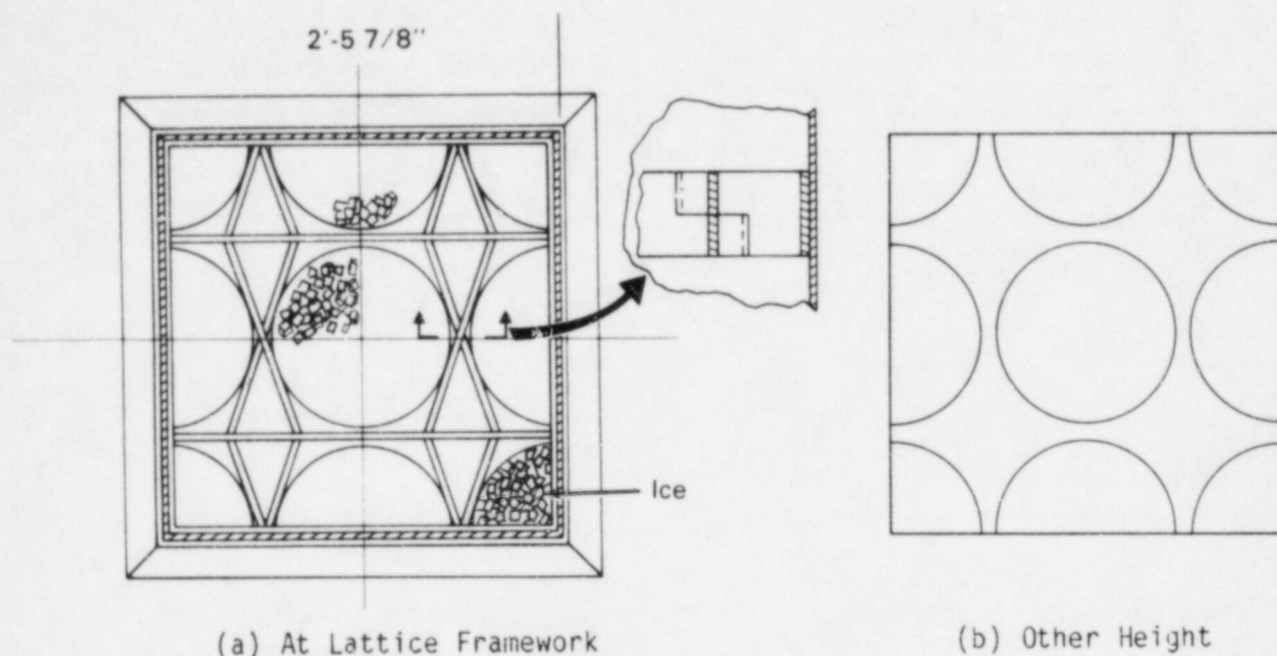


FIGURE 9. Test Section Cross Section.

Velocities at various locations within the test section will be measured by an insertion-type linear gas flow transmitter. Customized calibration and correction factors may be applied because of the wide variation in gas composition and temperature. The insertion probe will be manually positioned within the test section duct to measure local velocities, from which test section flow rates will be determined. Downward gas velocities are expected near ice surfaces, while the net gas flow rate is upward and decreases with height as the condensables (steam) are removed from the fluid stream. Gas velocities will also be measured using a flow visualization procedure. This procedure will employ a tracer (perhaps TiO_2 smoke) that will be released at the elevations of the window ports. The motion of the tracer will be recorded by a sophisticated video camera. Actual velocities will be derived from scale and time measurements.

Aerosol Generation. Aerosols with specific characteristics are required, including geometric median particle diameter, particle diameter geometric standard deviation (GSD), particle density, and aerosol mass and number concentration. While aerosol characteristics will be constant for the duration of each test, they will differ from test to test. The count median diameter (CMD) will be varied between 0.08 and 2.0 μm , the GSD will be approximately 2.0, and particle density will range from 1 to 4 g/cm^3 . Aerosol mass concentration will be controlled from values of less than 0.01 g/m^3 up to 20 g/m^3 ; this corresponds to particle number concentrations of $6(10)^3$ to $1(10)^6$ particles/ cm^3 .

Several techniques for producing polydisperse aerosols were investigated. The primary method of generating aerosols will be to disperse fine powders in a fluid energy mill. This technique is suitable for generation of all aerosols

except those with the smallest required CMDs. Particles suspended by the energy mill will be classified in the generator by a cyclonic separator. After the aerosol emitted from the generator is further classified, if necessary, to remove large particles, it will be ducted to the aerosol/gas mixing section. Aerosols with CMDs in the 0.08- μm size range will be generated using a quartz furnace or a plasma torch aerosol generation technique. The carrier gas will be nitrogen or argon. Output of the energy mill may be directed to the plasma torch to provide increased control over the production of these very small particles.

Materials (powders) currently being considered for aerosol generation include the following: cesium iodide, silver iodide, salt, tin, titanium dioxide, alumina, zinc sulfide, limestone, uranine, fluorescein, and anthracene. Other materials, such as magnesium oxide and manganese dioxide, are also being considered.

Test Design. Statistical experimental design techniques are being used to develop the test design, the combinations of parameters for which tests are to be conducted. Use of statistical design methods was deemed vital from the outset because of the large number of variables and expense of testing. Such methodology yields designs that maximize the information gained from the resources available. In addition, statistical methods provide assurance that unconscious or inadvertent biases do not affect the test design and limit the usefulness of resulting test data. The general approach used in developing the test design consists of four steps:

- List the thermohydraulic and aerosol variables of interest.
- Describe the experimental region of interest by: 1) specifying upper and lower bounds on the levels of the thermohydraulic and aerosol variables and 2) listing constraints that restrict the levels of the variables.
- Select the distinct test cases for the experimental design, including points on the boundary and in the interior of the experimental region of interest.
- Include replicate test cases in the design for the purpose of estimating the experimental error variance. The variability may depend on the location within the experimental region; thus, replicates at more than one test case are included in the design. These estimated variances are needed to statistically compare observed response values to ICEDF code-predicted values.

The list of thermohydraulic and aerosol variables and their upper and lower bounds were presented in Table 4. Although 11 variables are listed, three (gas pressure, initial ice fraction, and geometric standard deviation) are fixed. Thus, there are only eight variables of interest for experimental design development purposes. The upper and lower bounds on these eight variables define an eight-dimensional hyperrectangular region. Constraints among the variables modify the eight-dimensional hyperrectangle. Mathematically, the

thermohydraulic constraint cuts off several corners of the eight-dimensional hyperrectangle. In addition, the aerosol compatibility relationships also serve to reconfigure the experimental region to a seven-dimensional subspace. Thus, the experimental region of interest is a seven-dimensional subspace of the eight-dimensional hyperrectangle.

Experimental test cases from the boundary of the seven-dimensional experimental region were chosen by generating a candidate list of boundary points and then selecting test cases from the list using statistical techniques (see later in this section). The seven-dimensional experimental region has boundaries of dimension 0, 1, 2, ..., 6. The zero-dimensional boundaries are the vertices of the region, one-dimensional boundaries are edges connecting vertices, etc. Because the vertices are the most extreme boundary points, they comprise the first part of the candidate list. The one-dimensional boundary center points having a CMD at the midrange value of 0.6 μm are also included. (The CMD variable is expected to be the most important of the design variables, and hence, a middle level was chosen for study). The candidate set of boundary points from which boundary test cases were selected can be generated by taking all possible combinations of four sets of variable combinations: PS; FR; T, WF; and CMD, DEN, MC, and CC. The combinations were derived from upper and lower variable bounds (or upper, middle, and lower bounds in the case of CMD) and the constraints described above. Since the four sets of variable combinations contain 2, 2, 5, and 17 points, respectively, there are $2 \times 2 \times 5 \times 17 = 340$ zero- or one-dimensional boundary points (of the seven-dimensional experimental region) under consideration.

Based on cost considerations, it was determined that the minimum test design would consist of 15 test cases. The proposed characteristics of the 15 cases are shown in the following breakdown:

- 9 Experimental region boundary points
- 2 Experimental region interior points
- 3 Replicates of two boundary points and one interior point
- 1 Point outside the experimental region with no ice (IF = 0)
- 15

The nine-two split of the boundary/interior points was somewhat arbitrary. However, the considerable interest in validating the ICEDF code in the boundary areas of the experimental region led to locating the preponderance of the 11 at-large points on the boundary. For the limited number of tests that can be performed, boundary locations provide the most information about variable behavior within the experimental region of interest. Interior points are needed to provide information about the curvature of the response surface. The choice of both boundary and interior points satisfied the validation and calibration goals of the experiment.

The statistical computer program DETMAX (Mitchell 1974) was used to select several nine-point sets of boundary points from the candidate list. The nine-point sets resulted from DETMAX attempts to maximize (through the choice of the nine points) the determinant of the design matrix for the screening model

$$y = \beta_0 + \sum_{i=1}^8 \beta_i x_i,$$

where β values are constants and the x_i are the eight design variables T, FR, WF, PS, CMD, DEN, MC, and CC (see Table 4 for definition of design variables). The practical results of using DETMAX, its determinant maximizing criterion, and the above screening model are designs of a given size (nine points in this case) that are uniformly distributed over the experimental region boundary represented in the candidate list.

The current candidate fifteen-point test design (Table 5) is representative of the blending of statistical design and information on predicted aerosol deposition. DETMAX designs were reviewed with the following factors in mind. First, low steam fraction/small particle diameter combinations yield a low predicted aerosol decontamination factor (less meaningful experimentally because of expected experimental errors). Thus, the number of such cases should be limited. Second, the desired no-ice case parameter combination is low steam fraction, high volume flow rate, large and dense particles. Third, it is preferred that the no-ice case be repetitious of a full-ice case except that the initial ice fraction would be zero. In the most recent test design iteration, no changes were made to the DETMAX nine-point design as a result of concern over an excessive number of low aerosol deposition factor cases. Changes were made to the design to achieve the desired no-ice parameter combination. The resulting modified nine-point design serves as the core of the current fifteen-point design. Two interior points were specified by assignment of midrange parameter values. The balance of four cases is made up of three replicates and one no-ice case. Two replicates were selected from the nine-boundary points and the third is an interior point. It should be noted that the test design is still under review and changes are likely.

TABLE 5. Current Fifteen Volume Point Design

| Case No. | Temperature, °C | Soluble I or S | H ₂ O Mole Fraction | Vol. Flow, m ³ /s | Ice Fraction | Particle Density, g/cc | CMD, μm |
|-----------|-----------------|----------------|--------------------------------|------------------------------|--------------|------------------------|---------|
| 1 | 200 | I | 0.900 | 15 | 1 | 1.0 | 0.60 |
| 2 | 200 | I | 0.040 | 200 | 1 | 4.5 | 2.00 |
| 3 | 60 | S | 0.040 | 15 | 1 | 1.0 | 0.60 |
| 4 | 60 | I | 0.197 | 15 | 1 | 4.5 | 0.99 |
| 5 | 60 | I | 0.040 | 200 | 1 | 1.0 | 2.00 |
| 6 | 200 | S | 0.900 | 15 | 1 | 4.5 | 2.00 |
| 7 | 200 | S | 0.040 | 200 | 1 | 4.5 | 0.08 |
| 8 | 105 | I | 0.900 | 200 | 1 | 4.5 | 0.46 |
| 9 | 105 | S | 0.900 | 200 | 1 | 1.0 | 1.64 |
| 10 | 120 | S | 0.400 | 100 | 1 | 2.0 | 1.50 |
| 11 | 120 | I | 0.700 | 100 | 1 | 2.0 | 0.20 |
| Replicate | | | | | | | |
| 12 | 105 | S | 0.900 | 200 | 1 | 1.0 | 1.64 |
| 13 | 200 | S | 0.040 | 200 | 1 | 4.5 | 0.08 |
| 14 | 120 | I | 0.700 | 100 | 1 | 2.0 | 0.20 |
| No ice | | | | | | | |
| 15 | 200 | I | 0.040 | 200 | 0 | 4.5 | 2.00 |

| Case No. | MMD, μm | AMMD, μm | Mass Concentration, g/m ³ | Number Concentration, #/cm ³ |
|-----------|---------|----------|--------------------------------------|---|
| 1 | 2.54 | 2.54 | 0.98 | 1.000 × 10 ⁶ |
| 2 | 8.45 | 17.93 | 20.00 | 1.221 × 10 ⁵ |
| 3 | 2.54 | 2.54 | 0.01 | 1.018 × 10 ⁴ |
| 4 | 4.19 | 8.90 | 20.00 | 1.000 × 10 ⁶ |
| 5 | 8.45 | 8.45 | 20.00 | 5.495 × 10 ⁵ |
| 6 | 8.45 | 17.93 | 0.82 | 5.000 × 10 ³ |
| 7 | 0.34 | 0.72 | 0.01 | 9.998 × 10 ⁵ |
| 8 | 1.95 | 4.13 | 0.01 | 4.999 × 10 ³ |
| 9 | 6.92 | 6.92 | 20.00 | 1.000 × 10 ⁶ |
| 10 | 6.34 | 8.97 | 10.00 | 3.256 × 10 ⁵ |
| 11 | 0.85 | 1.20 | 0.05 | 6.869 × 10 ⁵ |
| Replicate | | | | |
| 12 | 6.92 | 6.92 | 20.00 | 1.000 × 10 ⁶ |
| 13 | 0.34 | 0.72 | 0.01 | 9.998 × 10 ⁵ |
| 14 | 0.85 | 1.20 | 0.05 | 6.869 × 10 ⁵ |
| No ice | | | | |
| 15 | 8.45 | 17.93 | 20.00 | 1.221 × 10 ⁵ |

FUTURE RESEARCH PLANS

Future plans for the laboratory tests include quantitative determination of the extent of particle retention associated with steam condensation on ice targets. Future efforts, however, will primarily be directed toward the engineering (unit-cell) tests. The preliminary facility and test designs will be

subjected to peer review. The current test design will also be reviewed for statistical and ICEDF validation merit. It is expected that these reviews will result in another iteration of the designs. It is anticipated that installation of the test facility will be completed during the second quarter of FY 1987 and that most of the tests will be completed by the end of the year.

At the conclusion of each test the data will be compiled and analyzed. The result of this analysis will be a complete definition of the test thermo-hydraulic conditions and aerosol characteristics. This will, in turn, lead to the decontamination factor for each engineering-scale test. Using this information, the ICEDF computer code will be operated and compared to the experimental results. Analysis of the code validation will not be complete until all tests have been performed. Most of the tests represent widely varying conditions and repetition of a few tests will be required to determine estimates of experimental precision.

Data reduction will be performed after each test. The expected data reductions are:

- temperature and pressure corrections to the flow measurements
- calculation of condensate generation rate, utilizing the load cell readings and sump pump operation history
- development of an overall heat transfer relationship incorporating steam composition and condensation information
- generation of time histories of temperature, gas composition, and ice inventory
- velocity measurements in the test section.

Estimates of the heat and mass transfer coefficients will be made based upon the temperature and steam concentration profiles. Zones of recirculation and general flow field behavior will be deduced from test section velocity measurements and videotaped tracer (smoke) motion. Ice melting shape and locations of fog will be obtained from videotapes and visual observations noted during the test. Global energy and mass balances will be calculated from the collected data.

Reduction of aerosol data will include determination of a water vapor concentration and bulk test section flow rate for each sampled elevation during each test, calculation of actual sample flow rates, and determination of changing particle characteristics at each sampling location.

Using the water vapor and tracer gas data, time histories of the gas composition and the net (upward) gas flow for each measurement station will be determined. Next, the actual sample flow rates for aerosol and gas measurements will be determined by comparing flowmeter data with calibration data at conditions of temperature and gas concentration similar to those that existed at the sample location during the sampling period. Measurements of selected

filters taken as water evaporation occurred following removal of the sample from the test environment will be plotted, and the data will be extrapolated to determine the fresh weight of the collected particulate mass. Laser transmission measurements will then be checked against the fresh weight of the analyzed filters. The aerosol mass flow rate time histories at each measurement station will then be calculated. These histories are obtained by multiplying the net gas flow rate by the ratio of the fresh (wet) particulate mass collected to the volume of gas sampled. Particle size distributions will next be plotted to determine aerodynamic mass median diameter (AMMD) and GSD. These results will be compared to determine change in particle size as a function of time and location in the test section. The effect of coagulation will be kept to a minimum by restricting the aerosol mass concentration input to the test section during testing; however, the effect of coagulation on particle size and particle number concentration will be estimated. A Lundgren sampler measurement of continuous particle size distribution at the inlet of the test system will provide a check on the stability of the generation process. Finally, the decontamination factor will be calculated by comparing the time history of aerosol mass flux into the test section to that of aerosol mass flux out of the test section. A global aerosol mass balance will be computed based on the time history of aerosol mass flux in and out of the test section; aerosol particulate concentrations in samples taken from the meltwater, rinse water, and scrubber; and knowledge of the generator efficiency and total aerosol mass introduced to the generator. Aerosol penetration of the ice will be estimated based on examination of ice cores taken immediately following testing.

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Contract Title: Effectiveness and Safety Aspects of Selected
Decontamination Methods FIN No. A6395

Contractor and Location: EG&G Idaho Incorporated
Idaho National Engineering Laboratory
P.O. Box 1625
Idaho Falls, Idaho 83415

Principal Investigators: S. W. Duce, F. B. Simpson, and J. W. Mandler

Objective: The initial objective in the 189 was to obtain information on decontamination methods that the NRC might be expected to review in the near future, (a) for effectiveness in reducing occupational dose and (b) for safety and integrity of components/systems following decontamination and restart. Since resolution of Generic Issue A-15^[1] the safety issue was deleted from the program objective. An itemized list of the objectives follows:

- Study current decontamination methods both processes and applications.
- Evaluate the current techniques used for determining decontamination factors (DFs) and make confirmatory and/or supplemental measurements.
- Evaluate the effectiveness of the studied decontamination methods to lower potential or actual personnel exposures.
- Report problems observed during the process application.
- Do follow-on studies of recontamination rates in previously decontaminated primary systems.
- Provide the NRC a data base for a regulatory position.

FY 1985 Work Scope:

TASK 1: Obtain and Evaluate Decontamination Data

a. Plant Instrumentation and Data

Obtain information from the plant, including description of the system(s) chemically decontaminated, chemical process used, and plant radiation survey data as well as plant sample analyses relevant to the decontamination campaign.

b. Observations

Observe the entire decontamination system and process operation. This includes preparatory work for decontamination, handling and mixing of chemicals, operation of the decontamination system, waste handling and waste processing. Operational problems and unexpected events will be assessed.

c. Measurements

Measure the gross radiation fields in reproducible geometries at various locations of interest in the system being decontaminated. The objective of these measurements is to provide confirmation of plant measurements and supplement these measurements where necessary. The specific locations for measurements and the times at which they are to be made will be selected in conjunction with the plant personnel. If technically practical, and plant personnel agree, limited gamma-ray spectral information will be obtained in order to determine specific nuclide decontamination factors. Thermoluminescent dosimeters (TLDs) will be used where applicable.

TASK 2: Assess Effectiveness of Successive Decontaminations

Evaluate the effect of successive decontaminations for those plants where an initial decontamination was performed prior to FY 1985. Evaluation will be of successive exposure reduction obtained, changes in operation and operating procedures, and waste handling.

TASK 3: Project Report

Write a draft project report concerning the entire scope of the program. This will include a summary of plant observations and measurements for each plant together with applicable information on the system decontaminated and the process application. The report will include recommendations for NRC research where generic overall research can be identified. Recommendations will also be made, if they can be identified, having potential for use in establishing a regulatory position (e.g., a Branch Technical Position).

TASK 4: Recontamination Rate Measurements

Make measurements, when available, at utilites after the first fuel cycle following a chemical decontamination of the primary recirculation system. These measurements will involve gamma-spectral measurements, TLDs, and hand held exposure instruments where applicable. These results will be used in evaluating the rates of recontamination following a chemical decontamination.

Summary of Research Progress:

TASK 1: Decontamination Observation and Measurement Trips

During FY 1985 EG&G personnel were able to observe chemical decontamination processes being applied at two different nuclear power electrical generating stations (Cooper Nuclear Station and Millstone Nuclear Power Station Unit No. 2). Data from a chemical

decontamination at the Robert E. Ginna Nuclear Station were also obtained. At each of the facilities visited a questionnaire was filled out that addressed the several different aspects of the decontamination process that were pertinent to this study. In addition to the observation of the process at Cooper Nuclear Station confirmatory measurements were made after the chemical decontamination. These measurements were gamma-spectral, TLDs, and exposure rate instrument readings.

With the addition of the utilities visited in FY 1984, a total of nine facilities were visited at the conclusion of the field studies, two pressurized water reactors for steam generator decontamination and seven boiling water reactors for primary coolant recirculation system and/or reactor water cleanup system decontamination. In addition to the nine facilities visited information on other chemical decontaminations were obtained from a total of four other facilities, one pressurized water reactor and three boiling water reactors. All of these facilities, with the exception of Dresden Unit No. 1, were commissioned between 1970 and 1975 and have been online for approximately nine to fifteen years. Four different chemical decontamination processes were observed and information on a fifth process was obtained.

The Cooper Nuclear Station chemical decontamination was performed by Pacific Nuclear Systems and Services Inc., using a PNS Dilute CITROX process. Pacific Nuclear personnel arrived onsite November 2, 1984, and equipment arrived from November 2 to 7. By November 14, 1984 all equipment had been assembled and leak tested and the decontamination process was initiated. The process proceeded smoothly and by 0200 on November 18, 1984 the decontamination process was completed. A total of 19.4 Ci of activity were removed with ^{60}Co accounting for approximately 77% of the activity removed from the pipe surfaces, with ^{54}Mn and ^{58}Co at 12.5 and 6.2%, respectively. A total of 250 ft^3 of resin was solidified for a final volume of 290 ft^3 . An average arithmetic mean DF of 40 was obtained in the primary recirculation system. The average DF was determined from the contact measurement results. This average DF resulted in an estimated savings of >450 man-rem and cost 14.2 man-rem to perform the chemical decontamination.

All measurements made by EG&G personnel at the Cooper Station were in good agreement with those made by RCT, the vendor hired by the utility to perform the before and after decontamination measurements.

The Millstone Nuclear Power Station Unit No. 2 chemical decontamination was also performed by Pacific Nuclear Systems and Services using PNS Dilute CITROX process. The chemical decontamination was performed following a chemical cleaning of the secondary side of both steam generators. The chemical decontamination of the steam generators was initiated April 30, 1985 and was completed on May 17, 1985. Material removed from one steam generator amounted to 69 Ci of which 70% and 25% were from ^{60}Co and ^{58}Co respectively. It was felt, by Pacific Nuclear personnel that a similar amount of activity was removed from the other steam generator, however the activity removed data

were not available at the time of writing. Waste volume was 235 ft³ of ion-exchange resins. The ion-exchange resins were dewatered in two high integrity containers, but not solidified as has been done at other facilities. The total buried volume of decontamination resin waste and liners was 300 ft³. Man-rem DFs for the steam generators were 6.4 and 6.0, and 8.5 and 5.5 for the hot and cold legs of the #1 and #2 steam generators, respectively. It was estimated that 1400 man-rem were saved due to the chemical decontamination.

TASK 2: Effectiveness of Sucessive Decontaminations

Data from the Millstone Nuclear Power Station Unit No. 2 steam generator chemical decontamination performed in 1983 by Combustion Engineering and Kraft Workers Union were obtained. These data were compared to the data obtained from the steam generator chemical decontamination performed in May of 1985 by Pacific Nuclear Systems and Services. The results indicate that the second chemical decontamination was more effective than the first, however what this finding means is clouded by the facts that each decontamination used a different process, a different vendor, and the application techniques were different. This task will have to be extended into FY 1986 and FY 1987 when more information concerning successive decontaminations can be obtained.

TASK 3: Draft Report

A draft report "Effectiveness and Safety Aspects of Selected Decontamination Processes" was written and sent to the NRC sponsor for review. Although the report was not transmitted until late October 1985 it is discussed here as the bulk of the report was finished during FY 1985 and was only delayed by delays caused in obtaining a few final reports from either licensees or vendors.

An appendix report was written for each facility visited and a copy was sent to both the licensee and the decontamination vendor for their review and comment. This practice was followed to allow either party to comment on the report and/or data prior to the report and associated data being used in the draft program report. These appendix reports described each plants experience with a chemical decontamination. Topics covered were (a) a brief description of the process as applied at the facility, (b) DFs obtained, (c) materials removed and waste generated, (d) man-rem savings, and (e) other pertinent information.

The draft report correlates all of the information gathered either from facility visits or from information obtained from licensees where chemical decontaminations were performed that were not observed. Table 1 is a summary of the recommendations and/or findings of the study. The following discussion will elaborate on each item listed in Table 1.

1. Decontamination Factors

Comparisons of the confirmatory measurements, made by EG&G, with the utility measurements show that the utilities' measurements are adequate, with the exception of the first

TABLE 1 SUMMARY OF RECOMMENDATIONS AND/OR FINDINGS

| Item | Recommendations and/or Findings |
|--------------------------------|--|
| Decontamination Factors | <ol style="list-style-type: none"> 1. Current Utility measurements are adequate. 2. TLDs can be used to determine the contact exposure rates. 3. Use of TLDs for contact measurements should be used as an ALARA method. |
| Reporting of an Average DF | <ol style="list-style-type: none"> 1. Use of a standard survey point map should be used in RWCS and PCRS chemical decontaminations. 2. Use a median value when reporting a system average DF. 3. Median DFs should be determined for similar components (i.e., risers, ringheader, etc.). Do not report a single system DF for PCRSs or RWCSs. 4. Identify the type of average DF (i.e., man-rem or Decon) reported. |
| Material Removed and Anomalies | <ol style="list-style-type: none"> 1. Total activity removed from all PCRSs and RWCSs was 517.6 Ci. 2. Reporting of ^{55}Fe was not always performed. 3. Chromium oxide concentrations are high in RWCSs. 4. A hard metallic fine with a high ^{51}Cr content has been observed in RWCSs and PCRSs. |
| Man-rem Savings | <ol style="list-style-type: none"> 1. Average man-rem savings per plant for all systems was 1480. 2. Man-rem expended per plant to perform the chemical decontamination and solidify the waste has been low, ~15 to 21 man-rem. |
| Waste Forms | <ol style="list-style-type: none"> 1. Most waste forms were solidified resins averaging ~150 ft³. |
| Lessons Learned | <ol style="list-style-type: none"> 1. Better QA is required for the vendor equipment and plant to facility interconnections. |

facilities visited, which had some problems with their measurements.

Supplementary measurements, using TLDs, were made to determine if these devices could be used to determine the before and after contact exposure rates. TLDs were placed at approximately 18 locations on the recirculation system piping and the results compared to measurements made by EG&G and facility personnel with exposure rate instruments. It was found that the TLD results were in good agreement with the instrument results (i.e., within 10%). An added benefit to the use of TLDs is the savings in dose to the health physics technicians performing the contact surveys. A TLD survey typically would require one-half to one man-hour in the drywell while the exposure rate survey required three to five man-hours.

2. Reporting of an Average DF

During the course of this study each of the decontamination vendors and/or the utilities used either different locations or number of locations in determining the DFs. These location DFs were then used to calculate an average DF. It was found that the results of the average DFs were erratic between vendors making comparison between various utility reported average DFs difficult. It was further found that the DFs reported by London Nuclear were more consistent partly due to their use of a standard survey map that was used at all facilities. When EG&G personnel put all the licensee data into a common format the job of making comparisons between the utilities was greatly simplified. Therefore, a standard survey point map should be used for all contact measurements made to determine an average DF in BWR recirculation systems and reactor water cleanup systems.

Not only were the vendors using different survey point locations they were also using different calculational techniques to determine the average DF. The different calculational techniques were found to result in average DFs, for the same data, being factors of ten different. After reviewing the data to determine if there was a better method for reducing the variance in the calculated average DFs EG&G personnel found that if an average DF was calculated by similar components the variation in the average DFs for a plant was reduced. It was also found that a median value, as calculated by numerically ranking the contact DFs and finding the central value, provided a more consistent and reliable technique for calculating an average DF by components.

One final point concerning the average DFs being reported is the type of average DF being discussed. It was found that there were two types of DFs being used, man-rem and Decon. A man-rem DF was an indication of the decrease in the general area exposure rates, where the Decon DF was an indication of the decrease in the pipe contact exposure rates. These two types of DFs were used by utility and vendor personnel without clearly stating which DF was being discussed. This was more prevalent when information was communicated between licensees or at meetings. With this type of confusion it is necessary that the type of DF being discussed be clarified.

3. Material Removed and Anomalies

Total activity removed from all BWR systems was 518 Ci, of which >75% was ^{60}Co . Total activity removed from steam generators was not determined due to lack of data.

There were three anomalies that were found during the course of this study. The first is in the lack of reporting of ^{55}Fe as a radionuclide removed. In two instances where ^{55}Fe was determined it proved to be a significant fraction of the total activity removed (i.e., >50%). The second anomaly was the large amount of chromium oxide found in the reactor water cleanup systems. In most cases poor DFs were obtained due to the chrome interfering with the process chemistry. An explanation as to why the chrome oxide is higher in this system than in the recirculation system is that the regenerative and non-regenerative heat exchangers cause such a large temperature change in solution that the chrome precipitates at these points. Therefore if a chemical decontamination of the reactor water cleanup system is to be performed the vendor should be prepared to deal with the high chrome oxide layer.

The last anomaly observed was reported by Peach Bottom^[2] personnel. During their chemical decontamination of 1984 a hard metallic fine with a high ^{51}Cr content was found in both the reactor water cleanup system and the recirculation system. No conclusion as to where the metallic fine came from was reached. At Monticello there was evidence that a fine, easily removable material was present in the reactor water cleanup system. No sample of the material was taken, but the behavior of the material in the cleanup system was similar to that noted at Peach Bottom.

4. Man-rem Savings

Average man-rem savings for all facilities and systems has been large, 1480 man-rem. A comparison of BWR system savings to PWR steam generator saving shows that the steam generators tend to have a higher average man-rem savings than the BWR systems (1690 compared to 1363 man-rem). The man-rem to perform the decontamination and solidify the wastes has been a small fraction of the savings, typically 15-21 man-rem. These results show that it is desirable to perform a chemical decontamination when a large amount of work is required to be

done on the primary coolant related systems.

5. Waste Forms

The typical waste form generated from a chemical decontamination has been solidified ion-exchange resins. The normal solidification matrix was cement/lime. The average volume of solidified waste was 150 ft³. There have been two instances where the waste forms were not solidified resins, one being the waste from Pilgrim Station where approximately 1300 gallons of decontamination solution was solidified using a cement/lime process. The other exception was the waste from the Millstone Unit No. 2 chemical decontamination where the ion-exchange resins were sluiced to two high integrity containers and then dewatered with no solidification.

6. Lessons Learned

There have been numerous lessons learned by both the utility and vendor personnel. The number one general problem area was dealing with the vendor equipment and/or the facility hookup. Most frequently, these problems were either related to the hose or the gasket seals on the hoses. Many of these problems could have been avoided with enforcement of proper quality control procedures.

TASK 4: Recontamination Rate Measurements

EG&G personnel traveled to the Quad Cities Nuclear Power Station Unit No. 2 for the purpose of making exposure rate and gamma-spectral measurements on the recirculation system piping. The purpose of these measurements were to evaluate the rate of recontamination of the primary coolant system piping following a chemical decontamination. The results of the EG&G and plant measurements indicate that the recirculation system experienced a 50% recontamination following one fuel cycle. Results of the gamma-spectral measurements have not been finalized. An evaluation of the effect on the surface concentration determinations due to the reactor coolant in the recirculation system piping needs to be performed.

Future Plans for FY 1986:

There are four tasks to be performed in FY 1986 (a) the finalization of the draft report, (b) make recontamination measurements at three facilities that were chemically decontaminated prior to the FY 1986 refueling outages, (c) obtain information on the effectiveness of successive chemical decontaminations, and (d) determine the effect of the aqueous radionuclide concentrations in the system piping on the surface concentration measurements. At the conclusion of the recontamination measurement phase information will have been obtained on the rates of recontamination in (a) a PWR steam generator, (b) in three BWR recirculation systems that were chemically decontaminated prior to an inservice inspection of the system welds, and (c) one BWR which had a complete recirculation system replacement. Depending on the findings of the measurement program and funding levels this work may be continued on into FY 1987 to obtain a larger data base on BWR systems where the recirculation system has been replaced.

References:

1. C. E. McCracken, "Regulatory Status Concerning Reactor Coolant System Decontamination and Steam Generator Chemical Cleaning," ANS Executive Conference Decontamination of Power Reactors, Springfield, Mass., September 16-19, 1984.
2. S. Nelson, Private Communications, Philadelphia Electric Company, May 1985.

CONTRACT TITLE: Decontamination Impacts on Solidification and Waste Disposal

PRINCIPAL INVESTIGATOR: Paul L. Picciolo

OBJECTIVE:

The objectives of the program at Brookhaven National Laboratory (BNL) are to identify the information and conduct the tests necessary to aid the NRC in making regulatory decisions on the solidification and waste disposal aspects of chemical decontamination processes. In particular, this program focuses on in-plant methods for converting decontamination wastes to more acceptable forms prior to disposal.

FY 1985 SCOPE:

In FY 1985, a laboratory evaluation of wet-air oxidation for degrading decontamination reagents was carried out. Additionally, a limited study of the solidification of the NS-1 decontamination reagent in Portland I cement was completed.

SUMMARY OF RESEARCH PROGRESS:

Earlier work in this program has provided an evaluation of potential decontamination processes, the wastes generated and potential waste management practices.⁽¹⁾ Subsequent laboratory studies⁽²⁾ have confirmed that incineration and acid digestion are capable of destroying selected decontamination reagents. Further, this work has assessed the solidification of simulated decontamination resin wastes in cement and vinyl ester-styrene.

The work described here was carried out in FY 1985, and further details can be found in Reference 3.

Wet-Air Oxidation of Decontamination Reagents

Wet-air oxidation has been applied to the treatment of several industrial streams.⁽⁴⁾ It was claimed that any combustible material that can be maintained in the liquid phase can be oxidized by this process. Wet-air oxidation has also been proposed for management of low-level combustible wastes and alpha-contaminated low-level wastes.^(5,6) A limited experimental program⁽⁷⁾ was conducted to determine the feasibility of using wet-air oxidation for volume-reducing spent ion-exchange resins prior to disposal.

Wet-air oxidation or pressurized aqueous combustion is a process that involves the oxidation of combustible materials in the presence of liquid. High pressures are employed to maintain the liquid phase. While some work has been reported on batch-type processes,^(5,6) the industrial units normally operate with a continuous feed. Recycling the residue and liquid effluent through the process may be advantageous to more complete degradation.

Experimental

The wet-air oxidation of decontamination reagents and ion-exchange resins was carried out in a batch mode using a 2-L Zipperclave supplied by Autoclave Engineers, Inc. A quartz insert was used to isolate the solution from the steel body. The apparatus was equipped with heaters, a cooling coil and a high speed stirrer. Parts on the autoclave head were fitted to allow liquid and gas sampling, and a thermocouple was used to monitor the solution temperature. Further details of the apparatus are given in Reference 3.

Samples of resins equilibrated with organic acids were prepared by mixing a measured quantity of ion-exchange resins with a solution of organic acid (or reagent) such that there was an excess of acid to exchange with the theoretical exchange capacity of resins. IONAC A-365 (Sybron) anion exchange resins were equilibrated with picolinic acid. This resin has a polyacrylic matrix with weak base exchange groups and was used because it is the resin of choice for the LOMI decontamination process reagent which uses picolinic acid. Quantities of the strong base anion exchange resin, IRN-78 (Rohm & Haas), were equilibrated with an excess of ethylenediaminetetraacetic acid (EDTA), citric acid, oxalic acid and LND 101A reagent. The amount of organic acid (reagent) on the resin/acid samples was determined by measuring the amount of acid in the solution after filtration. The carbon content of the samples was calculated based on the carbon contents of the resins and the acids.

The samples processed consisted of either organic acids, organic ion-exchange resins, or acid/resin mixtures, in one (1) liter of deionized water. Solutions contained either 0.5 or 1.0 percent by weight of material to be processed. Oxygen was used to pressurize the autoclave before heating the system. Typical ranges of operating conditions for processing the acid/resin mixtures were: initial oxygen pressure, 300 to 250 psi; maximum temperature, 210 to 225°C; maximum pressure, 550 to 720 psig; stirrer speed, 800 to 840 rpm; and run time, 5.5 to 6 h.

Aliquots of the processed solutions were collected at the ends of the runs and, in some cases, at different times during the runs. Liquid samples were analyzed for carbon content and, if applicable, the presence of organic acid. A gas sample was collected at the end of the process run time, after the system was cooled to room temperature, and analyzed by gas chromatography for permanent gases (H_2 , N_2 , O_2 , CO_2 , CO , and CH_4). Details of the analytical methods have been reported in Reference 3.

Results

A summary of the process results is given in Table 1. The amount of carbon initially present in the sample is based on the mass of the sample processed and the carbon content. Samples of resins and acid/resin mixtures have errors associated with the initial carbon which result from the determination of the carbon content of the samples.⁽⁸⁾

Values of percent organic carbon oxidized are used to monitor the efficiency of the process. These values are based on the amount of organic carbon measured in the process solution as compared with the initial amount of carbon present. All of the carbon in the resins or reagents was considered as organic carbon. Organic carbon remaining in solution after wet-air oxidation may be associated with a fraction of the original organic complexing agent which was not degraded by processing or with other hydrocarbons which were degradation products. If there is insufficient oxygen then hydrolysis can predominate oxidation^(4,9) during the wet-oxidation process. The products formed (low molecular weight carboxylic acids such as formic, acetic, propionic, etc.) can resist^(5,6,9) further oxidation. Williams⁽⁹⁾ and others have indicated that steady concentrations of formic and propionic acids are produced during the oxidation of butyric acid, and that acetic acid concentrations increase. The work done by Clark^(5,6) and by Bonnici⁽⁷⁾ indicates the production of acetic acid which was somewhat resistant to further oxidation. No detailed analyses were made of the organic chemicals remaining in the process solutions. However, when possible the post WAO samples were analyzed for the presence of the initial acid being processed. Based on the quantity of reagent remaining in solution, the percent reagent destroyed was calculated and the results are also listed in Table 1.

In initial testing, samples of Na₂EDTA and citric acid were processed for about 30 h. These runs were sampled after various times during processing and the data (although limited) suggest that oxidation of the organic carbon occurs relatively quickly and extended processing time does not result in a large increase in the amount of carbon oxidized. Figure 2.1 is a plot of reaction time vs fraction of organic carbon oxidized for the WAO of EDTA and citric acid. After 6.5 h, about 80% of the organic carbon in the EDTA sample was oxidized. An additional 10% of the organic carbon was oxidized after 30 h of processing. Oxidation of organic carbon in citric acid had essentially proceeded to a limiting value after 20 h. The heat was turned off at 28 h and no significant increase in carbon oxidized was observed. The sample collected at 45 h, after the system had returned to room temperature, showed <1% of additional carbon oxidized.

A striking difference between the results for EDTA and citric acid is the amount of inorganic carbon detected in the processed solutions (Table 1). Since the disodium salt of EDTA was processed, carbonates and bicarbonates are likely counterions for the remaining sodium present in solution. On the other hand, citric acid contains only C, O, and H, and dissolved CO₂ as carbonic acid may be the likely species to account for the inorganic carbon. The initial amount of Na₂EDTA contains 0.027 moles of sodium which would require an equal number of moles of HCO₃⁻ or half as many moles of CO₃⁻² for neutrality. Such quantities from the WAO would result in ≈320 mg and ≈160 mg of inorganic carbon in solution from bicarbonate and carbonate, respectively. The 280 mg of inorganic carbon may be the result of having approximately 75% of the sodium as NaHCO₃ and the remaining sodium as Na₂CO₃.

After processing the solutions containing EDTA and citric acid, the samples were analyzed for EDTA and citric acid remaining in solution. The results indicated that >99.9% of the EDTA and 99.8% of the citric acid were destroyed by processing. Other carbon containing species must remain in solution. These can include low molecular weight carboxylic acids such as formic, acetic, propionic, etc. (7,9)

Table 1
WAO Carbon Analyses of Post Run Liquid Samples

| Sample | Initial Carbon (mg) | Carbon Remaining ^a | | | Percent of Organic Carbon Oxidized ^b | Percent of Reagent Degraded ^c |
|--------------------------------------|---------------------|-------------------------------|----------------|--------------|---|--|
| | | Total (mg) | Inorganic (mg) | Organic (mg) | | |
| Na ₂ EDTA | 1616 | 440 | 280 | 160 | 90 | >99 |
| Citric Acid(CA) | 1875 | 360 | 20 | 340 | 82 | >99 |
| CA/Fe(NO ₃) ₃ | 1875 | 496 | 220 | 276 | 85 | N.D. |
| IRN-78 | 3360±20 | 1400 | N.D. | N.D. | 58 ^d | N.D. |
| IRN-78 | 1781±25 | 722 | 185 | 537 | 70 | N.D. |
| IONAC A-365 | 1569±40 | 743 | 299 | 444 | 71 | N.D. |
| EOC | 4968 | 1972 | 674 | 1298 | 56 ^f | 99,97,65 ^e |
| Picolinic Acid(PA) | 2570 | 1800 | 100 | 1700 | 34 | >99 |
| PA/IONAC A-365(1) | 2190±30 | 1230 | 402 | 828 | 62±2 | >99 |
| (2) | 2190±30 | 1227 | 303 | 924 | 58±2 | 97 |
| (3) | 2190±30 | 1160 | 325 | 835 | 62±2 | 95 |
| EOC/IRN-78 ^f (1) | 2360±30 | 1388 | 210 | 1178 | 50±1 | >99,99,94 |
| (2) | 2340±30 | 1011 | 90 | 921 | 61±1 | >99,99,92 |
| (3) | 2340±30 | 923 | 92 | 831 | 64±2 | >99,99,93 |
| LND 101A/IRN-78(1) | 2030±30 | 733 | 132 | 601 | 70±2 | |
| (2) | 2030±30 | 901 | 90 | 811 | 60±2 | |
| (3) | 2030±30 | 938 | 103 | 835 | 59±2 | |
| Blank | 0 | 10 | 0 | 10 | N.D. | N.D. |

^aOrganic carbon is determined from the difference between total carbon and inorganic carbon. N.D. means not determined.

^bPercent carbon oxidized is based on organic carbon remaining and the carbon initially present.

^cPercent degradation is based on the amount of organic acid left in solution compared with that initially present.

^dValue reported is based on total carbon and carbon available.

^eValues given are percent destruction of EDTA, OA, and CA, respectively.

^fSamples contained equimolar amounts (1.55×10^{-3} moles) of EDTA, OA, and CA.

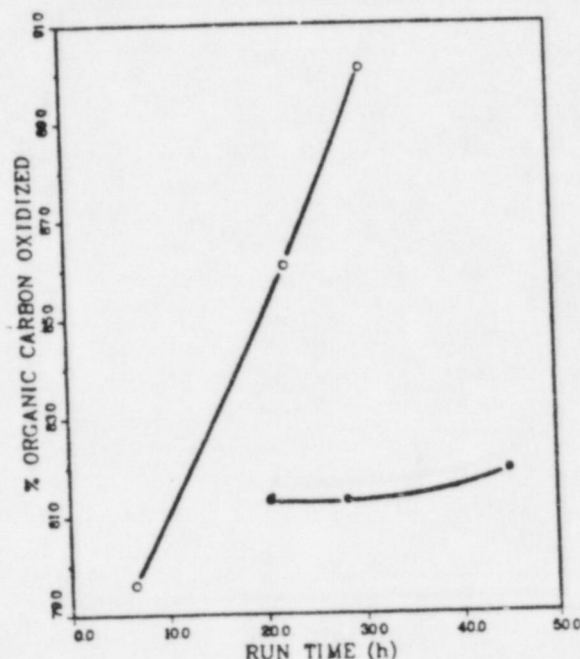


Figure 1. Percent organic carbon oxidized vs run time for the WAO of EDTA (o) and citric acid (.).

A 1-liter solution of 0.026M citric acid was oxidized for seven hours. The solution also contained 9×10^{-3} M ferric nitrate to determine if the presence of iron ions would enhance the amount of citric acid that is degraded. After processing, the final solution was an opaque red and there appeared to be about 85% oxidation of the acid, based on the amount of organic carbon remaining in solution. The presence of the metallic ion (Fe^{+3}) or perhaps decomposition of nitrate appeared to affect the oxidation. Ion chromatography data are not available for the $\text{CA}/\text{Fe}(\text{NO}_3)_3$ system to compare the amounts of CA left in solution with that determined previously; however, the amount of organic carbon remaining is smaller, indicating some increase in the oxidation of organic carbon.

WAO of the equimolar mixture of EDTA, citric acid and oxalic acid (1.5 weight percent in the simulated waste stream) appears to result in 56% oxidation of the mixture. This result is based on the amount of organic carbon left in solution. Ion chromatography data however, indicate that more than 95% of the EDTA and OA were oxidized compared to ~65% of the citric acid. The lower percent oxidation of citric acid in the EOC mixture from that observed for CA alone or in $\text{CA}/\text{Fe}(\text{NO}_3)_3$ may be due to the shorter reaction time, the absence of $\text{Fe}(\text{NO}_3)_3$ or because the sample was a mixture. It is possible that an unidentified product in the WAO process solution may interfere with the ion chromatography analysis for citric acid. A chemical having a retention time similar to that for citric acid would be quantified as citric

acid. A high estimate of the amount of citric acid in solution would give a corresponding low estimate of the amount of acid degraded. A complete analysis of the products in the process solution was beyond the scope of this work.

A 5.003-g sample of picolinic acid was processed for more than 6 h. The colorless solution recovered at the end of the run was slightly acid (pH = 6.3). Analysis indicated that only 34% of the organic carbon in the sample was oxidized but more than 99% of the PA was destroyed.

Based on the amount of organic carbon left in solution (Table 1), ion-exchange resins present at 0.5 percent by weight were oxidized $\approx 70\%$. No resin beads were observed in the post run solution although some "cloudiness" was noted in the solution. Comparing the two oxidations reported for IRN-78, the amount of resins used varied by a factor of two. If the percent oxidation is estimated from the total carbon remaining, there is no significant change (58% vs 60%). However, based on organic carbon in solution, run 2 showed 70% oxidation of the resins. In this run, a larger amount of oxygen was used.

The oxidation of the IONAC A-365 resins was comparable to that of IRN-78 resins when processed using similar parameters. The post run solutions were yellow in color. The pH of the solutions after processing were ≈ 4.4 for IRN-78 resins and ≈ 7.5 for IONAC resins. The oxidation products of the two resin types present different water chemistries after processing. These solutions may have different requirements for successful solidification of the process liquids prior to burial.

Replicate samples of PA/IONAC A-365 processed under similar conditions showed comparable percentages of carbon oxidized. The solutions recovered were pale yellow and neutral to slightly acidic. Small quantities of picolinic acid that were measured in the process solution indicated more than 95% destruction of the acid. Run 1, which was the largest of the three runs, showed $>99\%$ destruction of PA. The major differences in process parameters were a higher temperature, higher pressure, and higher stirrer speed.

Oxygen pressure was varied when processing samples of EOC/IRN-78. An increase of 40 psi in oxygen pressure resulted in an increase in the amount of organic carbon oxidized from 50% to more than 60%. Further, there was $\approx 50\%$ less inorganic carbon remaining in the process solutions of runs with the higher pressure compared with inorganic carbon in EOC/IRN-78(1). Analysis of the process solutions indicated that in all cases, $>99\%$ of EDTA, 99% of oxalic acid and about 93% of citric acid initially present in the samples was destroyed.

Three samples of LND-101A/IRN-78 were processed at similar temperatures and pressures but with different stirrer speeds. The resulting process solutions differed in that a pale yellow liquid was recovered from run 1 whereas the other process liquids were darker and contained a fine particulate. The carbon content of the process liquids showed that a greater percentage of organic carbon (70%) was oxidized in the first run than in subsequent runs ($\approx 60\%$). The last two samples were processed under similar conditions and had

similar data for carbon in solution. The major difference among the three runs was the higher stirrer speed of run one. Increased agitation will cause an increase in oxygen transport into the aqueous phase thus promoting oxidation.

A blank run was performed by processing 1-liter of deionized water under typical conditions used to process other samples. The results of the carbon analysis show that ≈ 10 mg of organic carbon were present at the end of the run. It is possible that the carbon was removed from parts of the apparatus that are difficult to clean between runs (e.g., pressure gauges, tubing), or was admitted to the system as contamination from the O-ring grease (Dow Corning high vacuum grease, which is a silicone lubricant, was used) or from the oxygen feed line. The 10 mg of carbon observed in the total carbon analysis of the solution is equal to between $\approx 0.5\%$ and 2.8% of the total carbon observed in the WAO runs of reagent samples. This small amount of carbon such as this is not likely to affect either the test being performed or the conclusions drawn.

Analyses of gas samples provide data on the various gas phase species produced during WAO of the simulated decontamination wastes. It was not intended in this work to correlate the carbon in the gas samples with the amount of organic acid destroyed. The analysis was limited to permanent gases (H_2 , O_2 , N_2 , CO , CO_2 and C_1 and C_2 hydrocarbons) by the choice of column used for the gas chromatograph. Thus, there are no data on the presence of nitrogen containing compounds like ammonia, volatile amines, and NO_x species which might be produced on the degradation of anion exchange resins and nitrogen containing organic acids (i.e., EDTA, picolinic acid).

Carbon dioxide was observed, as anticipated, in all samples since it is the expected oxidation product for carbon. The presence of CO , however, indicated that oxidation was not complete. Additionally, the presence of methane and hydrogen suggest that oxidation was not complete. These gases were observed in samples containing IRN-78 anion exchange resins and with citric acid.

Discussion

Based on the WAO process data, the percent oxidation of organic carbon in samples of ion-exchange resins equilibrated with reagents ranged between 50% and 70%. Samples of acids alone showed as much as 90% organic carbon oxidized. Analyses of the process solutions for residual organic acid indicated that nearly all of the original acid present was degraded. Citric acid appeared to be the most difficult acid to degrade of the four tested (EDTA, oxalic acid, citric acid, and picolinic acid).

The limited testing reported here indicates that oxidation is sensitive to the amount of oxygen present (initial pressure) and agitation. Process temperature and operating pressure also had some affect on the efficiency of degrading the materials. Extended processing times did not result in significant increases in the amount of carbon oxidized. The quantity of sample being

processed also limits the effectiveness of the treatment. These operating parameters should be considered in the design of a large-scale wet-air oxidation system.

Complete oxidation of organic carbon was not achieved under the process conditions used here. In particular, samples containing ion-exchange resins showed less than 70% oxidation of organic carbon. Even though the simulated wastes tested were not completely oxidized, wet-air oxidation was effective at degrading the decontamination reagents. Further processing of the waste may not be necessary if degradation of the organic complexing agents is the desired goal. However, the composition of the process solutions may need to be considered when planning the management of these wastes. For example, the presence of organic compounds may cause difficulties in direct solidification of the process liquids.

Based on the organic carbon analysis, process solutions contained organic species other than the original acid. Characterization of the process solutions was beyond the scope of this study, but it is likely that the composition may vary depending on the materials treated and the final degree of oxidation. Other workers⁽⁷⁾ have reported that the products of oxidation of ion-exchange resins contained $\approx 15\%$ low molecular weight carboxylic acid and that acetic acid, a major constituent, was somewhat resistant to further oxidation.

Consideration should be given to the organic acids remaining after wet-air oxidation regarding their potential to enhance radionuclide migration if released to the disposal environment. If the wet-air oxidation process were optimized, it is assumed acetic acid would be the major organic acid remaining. In general, acetate complexes are less stable than metal complexes of EDTA, or picolinic acid, which are used in decontamination reagents. Thus, the presence of acetic acid in the final waste form would be of less significance to the enhancement of radionuclide migration compared with the complexing agents used for decontamination (assuming comparable release of the acids from the waste form). If the decontamination waste was oxidized to an intermediate degree such that the decontamination reagents were degraded but acetic acid was not the only organic acid that remained, then the composition of the process waste should be evaluated to determine whether the treatment was of value. If oxidation was incomplete, it may be possible to treat the process liquid by filtration and ion-exchange to remove the radioactive species. The nonradioactive liquid presumably containing any complexing agents, could be handled separately from the radioactive waste.

Solidification of NS-1 Reagent in Cement

The waste generated by the chemical decontamination of a light water reactor will be disposed of in a shallow land burial site. Disposal of these wastes will be governed by the requirements established in the rule "Land Disposal of Radioactive Waste," 10 CFR Part 61, any future guidance given by the NRC and through site-specific criteria. The regulation establishes a

waste classification system for shallow land burial and a set of minimum performance objectives. NRC in the Technical Position on Waste Form provides guidance to waste generators on waste form test methods and results acceptable for demonstrating compliance with the waste stability criteria.

A laboratory evaluation of the capability of methods for solidifying decontamination resin wastes has been performed in order to assess whether they will meet applicable stability criteria. The primary objective of the solidification tests was to determine if cement and vinyl ester-styrene binders are viable agents for immobilizing simulated decontamination resin wastes. Based on current information, both mixed bed resins and anion exchange resins were selected for solidification testing. An acceptable waste form was one that met the requirements in 10 CFR Part 61 and the guidelines given in the Technical Position on Waste Form. Tests for acceptability included a visual inspection for the formation of a free standing monolith, the presence of free liquid, testing of the mechanical stability of the waste form and immersion in water. The details of this work and a discussion of the results can be found in Reference 2.

Work on the solidification of simulated decontamination wastes has been extended to include solidification of a selected liquid waste stream from a concentrated decontamination process. The results of the laboratory-scale tests that were performed are discussed in this section.

The NS-1 solvent, developed by Dow Chemical Company, is used for decontamination of BWRs. Although designed for use as a "concentrated" reagent, the NS-1 solvent has been used in a diluted form of 10% the original design concentration. Subsystems of various reactors have been treated with either dilute or concentrated NS-1.⁽¹⁰⁾

The waste stream expected from the use of NS-1 as a "concentrated" reagent was to be concentrated by evaporation of the process solvent. It was planned that the concentrated liquid would be solidified in vinyl ester-styrene. This was successfully demonstrated, and BNL reported⁽¹¹⁾ results of physical tests of the waste product.

Alternate methods of waste management can be used for handling the dilute NS-1 solvent. The reagents can be concentrated to a liquid stream, or removed from solution by ion-exchange. Both the concentrated liquid and resin wastes from a NS-1 process have been solidified in cement.⁽¹²⁾ Work in the present study was intended to evaluate the direct solidification of dilute NS-1 solvent in cement. Waste composites were prepared and examined for the presence of free liquid, tested for mechanical strength and tested for their ability to withstand immersion in water as recommended in the Technical Position on Waste Form.

A sample of NS-1 solvent was obtained from IT Nuclear Services which is licensed by Dow Chemical Company to apply NS-1. The composition of NS-1 is proprietary. NS-1 is an aqueous solution which in the 100% form contains

about 7.5% dissolved solids and has a pH of 3.6. The solvent contains chelates, complexing agents and inorganic salt plus a corrosion inhibitor. BNL received "spent" NS-1 (100%) which had three metallic compounds added to it: 1200 ppm ferric sulfate (or ferric citrate), 600 ppm nickel ammonium sulfate and 30 ppm copper sulfate.

It was believed that a probable waste stream from a NS-1 treatment would have a concentration approximately that of the 100% NS-1. This was based on knowledge that 100% NS-1 solvent can be used at the 10% dilution and then concentrated back to 100% prior to solidification. Further, BNL had a limited quantity of the material available for testing. It was anticipated, based on the results of the resin solidification tests, that pre-treatment by pH adjustment of the waste stream prior to solidification would be advantageous. Portland I cement was chosen for the solidification of NS-1. Details of the solidification are given in Reference 3.

The compressive strengths of the cement forms were measured according to the ASTM C39-80 test method. The average compressive strengths of four NS-1 samples and their controls are given in Table 2. Although the mechanical strengths of the controls were smaller than those of the forms containing NS-1, all forms had compressive strengths larger than the 50 psi recommended in the TP on Waste Form.

Table 2
Compressive Strengths of Cement Composites

| Sample | Cure Period (days) | Compressive Strength (psi) | |
|---------|-----------------------|----------------------------|----------------|
| | | pre-immersion | post immersion |
| NS-1 | 34 | 5840+240 | 6050+560 |
| Control | 31 | 4370+240 | 4870+100 |

Four NS-1 composites and three controls were immersed in 1-L of water after a 28-day cure period. After the 90-day immersion test, there was no visible sample deterioration, and the forms had compressive strengths greater than the 50 psi recommended in the TP on Waste Form.

These limited tests demonstrate that NS-1 can be solidified in cement to at least meet the Class A waste form requirements. However, assuming that half of the 7.5% solids in NS-1 are chelating agents then the waste (NS-1 plus water) solidified in these tests contained less than 3% chelating agents. In the field, it may be desirable to concentrate the solvent to a waste stream containing a higher concentration of dissolved solids and thus a higher concentration of organic reagents. Solidification of these wastes may not proceed as it did in the tests reported here. As with the work done on resin

wastes, it is expected that variations in the composition of the liquid decontamination waste stream must be taken into account when establishing formulations for solidification.

Summary

Wet-air oxidation appears to be an effective method for degrading simulated decontamination wastes. The process data given in this report show that even though the amount of oxidation of total organic carbon in the acid/resin mixtures ranged between 50 and 70%, there was greater than 92% degradation of the organic reagents. The percent degradation of the organic reagents is based on the quantity of reagent present after processing compared with the initial amount in the sample. It is believed, based on data collected in the ongoing BNL study, that WAO can be comparable to incineration and acid digestion for degrading decontamination wastes. Testing in this program was limited to three simulated waste streams: reagents alone, anion exchange resins and anion exchange resins equilibrated with reagents. For each of the processes evaluated in this study, only limited efforts were made to optimize the process apparatus or operating parameters. It is recommended that waste-specific operating parameters be established if any of these processes are to be used commercially.

The acid/resin mixtures tested in this program were intended to simulate dilute process decontamination wastes. Resin wastes from actual reactor system treatments can include cation exchange resins, metal ions and radionuclides. The presence of these materials in the waste stream may affect the efficiency of degradation by wet-air oxidation. Although it was indicated in earlier studies that transition metal ions may catalyze the oxidation of organic materials (Ca^{+2} , Fe^{+2} , and Mn^{+2} nitrates were used as a catalyst by Clark, W. E.⁽⁵⁾), the presence of iron in processing citric acid had a limited effect. It would be necessary to determine whether a catalyst will increase oxidation of the resins at lower operating temperatures.

The present data suggest that the degradation of each acid can be affected by process parameters such as temperature, pressure, and agitation. Agitation (stirring) and oxygen overpressure did increase the amount of organic carbon oxidized in tests of EOC/IRN-78 and LND-101A/IRN-78. An increase in process temperature resulted in an increase in the degradation of picolinic acid in samples of PA/IONAC A-365. However, the temperature increase had a corresponding increase in operating pressure which promotes the rate of oxygen transfer into solution. It is not known if an increase in pressure alone would have given a similar result. Analyses of process solutions showed $\approx 99\%$ degradation of EDTA and oxalic acid, but a somewhat lower percentage degradation was observed for picolinic acid ($>95\%$) and citric acid ($\approx 93\%$) when processed under similar conditions. Thus, the composition of the waste stream to be processed should be considered when establishing process parameters.

The size of the batch-type processor used in the BNL study limited the sample size that could be treated. Additionally, limitations in the pressurization equipment limited the amount of oxygen in the system. Thus, only

samples of 0.5 weight percent of waste-to-water were processed which is below the 2 to 20 weight percent organic matter expected to be treated by WAO. Recycling the process solution or use of a flow system may increase the total degradation of the waste. The design of the WAO reactor must consider the concentration of waste to be processed; in turn, this will affect the effluent streams (off-gas and liquids).

Management of the effluent streams from the WAO process will need to be considered. The process solutions will contain suspended solids, inorganic salts, and can include various inorganic and organic carbon-containing species. Further, the data show that the pH of the liquid stream is dependent upon the material processed. It is anticipated that the volume of the liquid stream would be reduced prior to any further treatment. If this waste stream were to be solidified prior to disposal in shallow land burial, then it should be demonstrated that the subsequent product meets the appropriate requirements of 10 CFR Part 61.

The off-gas stream from the wet-air oxidation of decontamination wastes will contain CO_2 , CO, and water vapor. However, methane and hydrogen have been observed and nitrogen-containing gases such as volatile amines and NO_x gases may also be present. Thus, off-gas treatment may be necessary.

Limited tests have demonstrated that a liquid waste stream from a decontamination using NS-1 reagent can be solidified in cement after pre-treatment by pH adjustment. The test reported here was for a single concentration of NS-1 in the waste stream. It is anticipated that the concentration used in this test is low compared to what may be used in full-scale solidifications under field conditions. As with the work done on resin wastes, it is expected that variations in the composition of the liquid decontamination waste stream will have to be considered when establishing process parameters for solidification.

FUTURE RESEARCH PLANS:

FY 1986 research will include an evaluation of bitumen as a solidification medium for decontamination wastes and the impacts of decontamination wastes on standard containers. The objective of the work is to generate data that will aid NRC in assessing problems that may be encountered with the long-term storage or disposal of decontamination wastes and to evaluate the acceptability of on-site decontamination waste handling and solidification processes. These experiments would investigate the corrosion or degradation of container materials in contact with decontamination resin wastes and include examination of the effects of irradiation on decontamination wastes which may result in gas generation or the release of liquids.

It is also believed that the biodegradation of decontamination wastes should be studied, as well as the effects of thermal cycling on solidified wastes. The tests planned are consistent with those recommended by NRC to demonstrate waste form stability.

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FISCAL YEAR 1985 SUMMARY REPORT

TITLE: Evaluation of Nuclear Facility Decommissioning Projects (ENFDP) Program

PRINCIPAL INVESTIGATOR: R. L. Miller and K. S. Scotti
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OBJECTIVE:

The objective of the Evaluation of Nuclear Facility Decommissioning Projects (ENFDP) Program is to provide the Nuclear Regulatory Commission (NRC) staff with data which will allow an assessment of man-hours expended, radioactive wastes generated by type and volume, alternative methods of decommissioning and occupational doses incurred during decommissioning activities. The data provided will also include available cost information to assist the NRC in determining the proper amount of funds which must be available to ensure timely and safe decommissioning of facilities.

FISCAL YEAR 1985 WORK SCOPE

The work effort for FY 1985 was divided into six categories for proper activity definition and cost accounting. Those categories are:

- Program Management
- Identification of Facilities
- Collection of Data
- Analysis and Reporting
- Comparison Studies
- International Nuclear Reactor Decommissioning Planning Conference

For a detailed description of the overall program plan and definition of the activities under the first five categories, see NUREG/CR-2522, Rev 1, "Evaluation of Nuclear Facility Decommissioning Projects, Program Plan," December 1983.

During FY 1985, UNC was also responsible for conducting an international conference on nuclear reactor decommissioning planning for the NRC.

Specific tasks and objectives established for FY 1985 were:

Program Management

- Publish annual status report for FY 1984 ongoing projects by January 1, 1985.
- Revise ENFDP Program Plan as required.
- Prepare monthly summary reports which will present, as a minimum: the progress during the period; work planned during the next period; budgeted and actual costs by task; milestone status; and cost/milestone variances.

Identification of Facilities

- Identify additional facilities for data collection in FY 1986/1987 (continuing effort). Establish working agreements with licensees/owners of the facilities which will be evaluated in Fiscal Year 1986.

Collection of Data

- Collect data at Shippingport, TMI-2, Humboldt Bay and one other facility to be identified.

Analysis and Reporting

- Submit a final report on TMI-2 polar crane recovery to the NRC for publication by December 1, 1984.
- Prepare a draft report on TMI-2 activities up to the time of head removal. Transmit to the NRC two months after head lift begins.
- Issue Project Summary Report for Humboldt Bay decommissioning (contingent upon on-time decommissioning).

Comparison Studies

- Maintain and update computer software to support comparison studies (continuing effort).

International Nuclear Reactor Decommissioning Planning Conference

- Prepare for and conduct an international conference for the NRC on nuclear power reactor decommissioning in July 1985. Prepare proceedings for distribution by September 30, 1985.

SUMMARY OF RESEARCH PROGRESS

Program Management

Program Management includes those tasks involved in preparing the Program Plan, as well as routine management, project control, and reporting.

The Annual Summary Report for Fiscal Year 1984 was prepared for the NRC and published in January 1985 (Reference 1). Included as appendices were draft reports of data from TMI-2 recovery efforts and Shippingport Atomic Power Station (SAPS) decommissioning. The TMI-2 draft report covered radioactive solid waste shipments and the Auxiliary and Fuel Handling Building (AFHB) decontamination. The SAPS draft report covered data entered in the decommissioning data system (DDS) during the fiscal year from information derived from activity specifications as revised in the SAPS decommissioning plan. This preliminary information, divided into multiple categories, will become the specific report structure for SAPS decommissioning data as the work is initiated and completed.

The ENFDP Program Plan (Reference 2) was reviewed. As there were no significant changes, the document was not revised or reissued.

Monthly summary reports covering work progress and costs were provided to the NRC Program Manager. Total program expenditures through the end of FY 1985 were \$1,518,000. Actual cumulative costs of each of the six program work categories for FY 1985 were:

PROGRAM COSTS FOR FISCAL YEAR 1985

| <u>Task</u> | <u>FY 1985 COST</u> <u>(\$000'S)</u> |
|-------------------------------------|---|
| Program Management | \$ 92 |
| Identification of Facilities | 9 |
| Collection of Data | 46 |
| Analysis and Reporting | 156 |
| Comparison Studies | 27 |
| Decommissioning Planning Conference | <u>104</u> |
| TOTAL | \$434 |

Identification of Facilities

Identification of facilities entails the selection of appropriate reactor facilities for data collection during future years. The major constraint for this activity is the availability of significant decommissioning projects that can provide relevant data for the program.

Four additional reactor decommissioning projects were identified for inclusion in the program in FY 1986/1987. Three of the projects are located in the Federal Republic of Germany (Lingen, Gundremmingen, and Niederaichbach). A TRIGA Mark IV research reactor owned by the Northrop Corporation in Los Angeles, California, was the fourth addition.

A working agreement was established to collect project data from the German reactors through an existing international technology exchange agreement between the U. S. Department of Energy and the Federal Ministry for Research and Technology of the Federal Republic of Germany (BMFT-FRG). The Lingen and Gundremmingen reactors do not fall under the direction of the BMFT-FRG. However, the utilities involved have agreed to cooperate in the ENFDP program. By making use of the existing U.S.D.O.E./BMFT-FRG agreement, many months of negotiating time and ENFDP program funding were saved.

Data collection from the Northrop TRIGA reactor decommissioning will be by direct purchase order with the dismantling contractor, Chem-Nuclear Systems, Inc.

Lingen Reactor

The KWL Nuclear Power Station (Lingen) is a boiling water reactor with a thermal output of 520 MW. Construction began in 1964 and full power operation was initiated in 1968. The plant operated for 10 years with an average availability of 60 percent and a total output of 11 TWh. In January 1977 the plant was shut down to check the heat exchangers. During the investigation small cracks were found and

replacement of the heat exchangers was defined as an essential modification of the plant which would require a new licensing procedure.

Because of the heat exchanger cracks, other backfitting needs with an estimated cost of 100 to 200 million DM (approximately \$42 and \$84 million U. S.), and no guarantee of receiving a new operating license, the plant was permanently shut down.

Decommissioning plans are to place the facility in safe storage then wait another 25 years before demolishing the structures. Collection of data will concentrate on decontamination and monitoring techniques, and radionuclide inventories completed to date. No further characterization is planned until just prior to demolition.

The safe storage/demolition method of decommissioning planned for the Lingen reactor is similar to the method proposed for the U. S. Indian Point-1, Dresden-1 power reactors and the University of Texas and Georgia Tech research reactors.

Niederaichbach Reactor

The KKN Niederaichbach reactor (Landshut, Bavaria) is a heavy water moderated, CO₂ cooled pressure-tube reactor designed for a nominal output of 100 MWe. Construction began in 1966 and was completed in 1972. From 1972 to 1974 the plant operated for 18 full-power days with 40 percent of its nominal power. For economic and technical reasons, the plant was permanently shut down in 1974.

Planned decommissioning is in two phases. The first, safe storage, was completed in 1978. The second phase will restore the site to "Green Field" condition - release for unrestricted use with the land being returned to green field appearance. Licensing to begin this phase is expected in 1986 and the second phase activities are expected to take six to seven years.

Gundremmingen Reactor

The KRB Gundremmingen reactor (between Ulm and Augsburg in Bavaria) is a 800 MWt boiling water reactor of the GE BWR design concept which operated for about 11 years. Construction began in 1962 and was completed in 1966. The plant was permanently shut down in 1977 following a short-circuit in the grid which resulted in damage to the plant and contamination in the turbine hall. During the years of operation, the plant generated 15 billion kilowatt hours of electric power with an average availability of 75 percent.

Decommissioning plans call for safe storage of the reactor and auxiliary buildings and partial dismantlement of selected systems in the turbine hall. The intent of this plan was to reduce radioactive waste volume as much as possible (by at least a factor of 10) and to reclaim usable materials. Parts have been removed from the turbine hall including the electric generator.

The Gundremmingen reactor is similar in design to the U. S. Dresden-1 reactor. Data collection at this facility will add to existing knowledge and understanding of the time, manpower and exposure factors involved in partial dismantlement/partial safe storage of similar design BWRs.

Additionally, the German decommissioning work is expected to make substantial use of remote tooling which is of particular value in applying ALARA techniques and in assessing the impact of use of ALARA on overall decommissioning costs.

Because of the types of activities planned for the FRG reactors, collection of data will be of particular value in augmenting the decommissioning data base for BWR reactors and for U. S. plants considering the SAFSTOR decommissioning mode. This planned data collection activity and subsequent analysis is in accordance with the overall ENFDP program plan and philosophy.

Northrop TRIGA

The Northrop TRIGA facility (located in Hawthorne, California) was a 1 MWt research reactor capable of pulsed operation to 1800 MWt. Operation began in 1962 and continued until 1982. Decommissioning planning was initiated in 1983 and decommissioning operations began early in 1985.

Planned decommissioning is to fully dismantle portions of the facility and release the structure for unrestricted use. Many of the dismantling tasks scheduled for the TRIGA facility are comparable to the work performed at the North Carolina State University research reactor and the Ames Laboratory research reactor. Addition of the Northrop TRIGA data to the ENFDP database will provide sufficient information to begin comparison studies for the dismantlement of research-type reactors.

Collection of Data

The Collection of Data task includes site data collection, subcontracts or agreements with licensees, and the search and recording of reference published data.

Shippingport

Actual decommissioning work at the Shippingport Atomic Power Station (SAPS) did not begin until September 17, 1985. An initial site visit was made during the second quarter of FY 1985 to establish communication channels and to explain the ENFDP program to SAPS decommissioning project management.

During the fourth quarter of FY 1985, site radiological characterization data were obtained from the decommissioning operations contractor, General Electric Company.

Activities at Shippingport during the majority of the fiscal year concentrated on preparations for the start of physical decommissioning culminating in an Operational Readiness Review (ORR) in August 1985.

The Shippingport Atomic Power Station decommissioning project is a major activity of the U. S. Department of Energy's (DOE) Surplus Facilities Management Program (SFMP). Shippingport will be decontaminated and then dismantled.

The Shippingport reactor is a PWR designed to produce 72 MWe. It was the first plant of such size in the world that was operated solely to produce electric power. It began operation in 1957 and was shut down in October 1982. Over its life, the plant produced 6.35 billion kilowatt hours of electricity.

The SAPS planning, dismantlement and removal procedures are being conducted by the U. S. DOE as a demonstration project for the application of techniques useful for decommissioning commercial reactors.

While Shippingport's 72 MWe power rating is much lower than that of modern plants, it is physically nearly as big, due to its early design. Information relating to decontamination, dismantling and removal of many of the reactor's systems can provide useful information in planning for future dismantlement of facilities currently in SAFSTOR or being considered for near term decommissioning.

The Humboldt Bay reactor, currently being put into SAFSTOR, is a BWR reactor of similar power rating. Data gathered at Shippingport will also be useful in final decommissioning planning for the Indian Point-1, Peach Bottom-1, Enrico Fermi-1, and Dresden-1 reactors.

Three Mile Island

Data collection at the TMI-2 site was ongoing throughout the year. The data obtained from TMI-2 has been divided into nine categories to correspond to the nine major tasks identified for the facility's recovery efforts. The categories are:

- Reactor Coolant System and Systems Decontamination
- Reactor Building Decontamination
- Reactor Defueling and Disassembly
- Auxiliary and Fuel Handling Building Decontamination
- Common Support Facilities and Systems Operation
- Plant Stability and Safety Activities
- Liquid Waste Handling
- Solid Waste Handling
- Radioactive Waste and Laundry Shipment

Data collection at TMI-2 during FY 1985 concentrated on reactor building decontamination and reactor defueling preparation. Data collection for system decontamination and waste shipment were also included during the period.

Four NUREG documents were prepared for publication to provide summary status reports of TMI-2 activities (References 3 through 6). Those draft documents summarize the following:

- Radiation exposures, manpower and time spent in radiation areas during the performance of tasks related to decontamination and repair of the Reactor Coolant System (RCS) and other reactor systems (Reference 3).
- Radiation exposures, manpower and time spent in radiation areas during the performance of tasks related to decontamination of the reactor building (Reference 4).
- Radiation exposures, manpower and time spent in radiation areas during the performance of tasks related to preparation for reactor defueling and disassembly (Reference 5).
- Waste disposal site locations, dose rates, curie content, waste description, container type and number, volumes and weights of radioactive waste and laundry shipments from TMI-2 since March 1979 (Reference 6).

Humboldt Bay

Data collection at the Humboldt Bay facility was conducted on an approximately quarterly basis. During FY 1985 data was obtained on radiological characterization of the site, personnel exposures by task and job function, and radioactive waste shipment and disposal.

The estimated man-hours and man-Rem exposures were obtained and input into the decommissioning data system (DDS). These estimates are subject to change since the utility's plan to place the facility in a 30-year safe storage condition is awaiting NRC approval.

A NUREG document was prepared for publication to provide a summary status report for Humboldt Bay (Reference 7). It includes information collected from the facility decommissioning plan, environmental report, and other sources made available by the licensee. This information has also been included in the decommissioning data system.

Northrop TRIGA

Data collection at the Northrop TRIGA reactor dismantling was initiated during the third quarter of FY 1985. An agreement was completed with the decommissioning contractor, Chem-Nuclear Systems, to acquire the desired program information upon completion of the facility dismantling. This data will be analyzed and input into the DDS following receipt. Receipt of data is expected during the third quarter of FY 1986.

Analysis and Reporting

The Analysis and Reporting task includes development of a computer program to store and allow manipulation of decommissioning data, development of a standardized reporting format, compilation of site data, and issuance of facility decommissioning reports.

Three Mile Island

A NUREG final report on recovery of the TMI-2 polar crane was prepared for publication (Reference 8). The report contains information on radiation exposure, manpower by craft, and time spent on repair of the crane. The report covers the time period from entry number 10 on May 14, 1981, "Initial Climb to Polar Crane with Safety Equipment" until full mobility and operations were restored during entry number 311 on December 19, 1983. The report was published in August 1984 - five months ahead of the scheduled ENFDP milestone.

A draft report on TMI-2 activities up to vessel head removal was originally scheduled as part of the ENFDP fiscal year 1985 activities. Reactor vessel head removal activities followed directly from the polar crane recovery efforts. As the data related to vessel head removal was included in the report prepared on reactor defueling and disassembly (Reference 5), a separate report was not prepared. Included in Reference 5 are the radiation exposure, manpower by craft, and time spent on each activity from control rod drive mechanism removal, thermal insulation removal, detensioning of vessel head stud bolts, and other related work up to final removal of the vessel head.

Humboldt Bay

The Humboldt Bay safe storage preparations were originally planned to be completed by June 1985 with final shipment of waste by December 31, 1985. Currently the schedule has slipped to completion of safe storage preparations by June 30, 1986. As summary data was not available, a project summary report for Humboldt Bay was not completed during FY 1985. However, a status report draft was prepared for publication (Reference 7). The report gives the current status of preparations to place the facility in SAFSTOR and includes information collected from the facility decommissioning plan, environmental report, and other sources made available by the licensee. This information was also included in the decommissioning data system in preparation for data analysis and summary.

Comparisons/Studies

Comparison Study activities include: comparison of site specific data with information provided in the reference NRC document for that type of facility; comparison of relative costs and decommissioning techniques used at each site; development of a system that can be used by the NRC to evaluate future licensee's decommissioning plans for adequacy, based upon the field experience gained during this program; and issuance of summary reports that will discuss the data collected in the field, the relationship to the assumptions identified in the NRC decommissioning studies, the relative value of various decommissioning techniques based on actual experience, and the system for assessment of future decommissioning plans.

Comparison studies can be made after decommissioning data have been obtained from a reasonable number of facilities of each type (PWR, BWR, Research and Test Reactors). When the data base is large enough to permit

validation of data points, comparisons can then be made between the experiences at these facilities and with the NRC studies pertaining to decommissioning.

Initiation of summary comparison studies was again delayed due to lack of enough data for any one reactor type in the data base to begin this work. Work was ongoing throughout the year to prepare the system to perform detailed comparison studies as soon as the data base contains enough information to make those analyses meaningful. Work continued on streamlining the interface between the PC terminals and the main frame computer and on preparations for call-in capability to access the Decommissioning Data System (DDS).

As a routine annual task, the data base cost escalator feature was updated using the Consumer Price Index (CPI) figures received in early 1985.

Information currently in the DDS concerns the following reactors:

- . Elk River Power Reactor
- . Shippingport Atomic Power Station (engineer estimates)
- . Plum Brook-1 Test Reactor
- . Ames Laboratory Research Reactor
- . North Carolina State University Research Reactor
- . Enrico Fermi-1 Power Reactor
- . Reference BWR
- . Reference PWR
- . Reference Test Reactor
- . Reference Research Reactor
- . Three Mile Island-Unit 2 Power Reactor
(extensive data on recovery activities)

International Nuclear Reactor Decommissioning Planning Conference

During the fourth quarter of FY 1984, UNC was directed to begin preparations for an NRC workshop on decommissioning planning for nuclear power reactors to be held in July 1985.

The objective of the workshop was to provide an international forum for the exchange of philosophy, ideas, and technology information on planning for decommissioning. Papers were invited for submission in the following areas:

- Philosophy and Guidance
- Funding
- Decommissioning Plans and Experience
- Management of Decommissioning Wastes
- Engineering Problems

As planning progressed, sufficient interest in participation was evident to increase the scope from a workshop of 50-100 attendees to a conference capable of accommodating up to 250 participants.

The conference was held July 16-18, 1985, in Bethesda, Maryland. Actual conference attendance was 186 persons including 20 guests. Of the total number of participants, 43 were from countries outside of the United

States. Countries represented were: Austria, Belgium, Canada, Federal Republic of Germany, France, Israel, Italy, Japan, Sweden, Taiwan, the United Kingdom and the United States.

The technical program included 37 invited and contributed papers from 10 countries, the International Atomic Energy Agency (IAEA), and the Commission of European Communities (CEC).

Two page summaries of papers presented at the conference were published and distributed upon registration. Proceedings with copies of the full papers, a transcription of the guest speaker's (Dr. Dixy Lee Ray) presentation, and the keynote address by Mr. G. A. Arlotto, Director of the Division of Engineering Technology of the NRC, were prepared in camera-ready form and sent to the NRC Program Manager on September 19, 1985, for publication as NUREG/CP-0068, two weeks ahead of schedule.

All presenters and session chairmen were photographed during the conference. Following the conference these photographs were compiled into a guidebook which also contained the name, address, company affiliation, country, and telephone and/or telex numbers. Copies of the guidebooks were distributed to the U. S. NRC Program Manager, the UNC Decommissioning Programs director and the UNC Decommissioning Programs Department Technical Library. One copy was retained for ENFDP program use.

A final programmatic report (Reference 9) and a financial summary report (Reference 10) were prepared and forwarded to the NRC Program Manager for information on September 30, 1985.

FUTURE RESEARCH PLANS

The ENFDP program will continue through fiscal year 1986. The work effort for FY 1986 will continue to be divided into the five activity categories of program management, identification of facilities, collection of data, analysis and reporting, and comparison studies.

Specific tasks for FY 1986, subject to final budget approval, will be:

- Draft year-end progress report to the NRC.
- Prepare an annual summary report for the program for NRC publication.
- Review and revise the ENFDP Program Plan as required.
- Identify additional reactor facilities being decommissioned (or having information relevant to decommissioning) or scheduled for decommissioning in the 1986-1987 time frame as candidates for inclusion in the 1987-1988 ENFDP program.

Identified facilities will be evaluated and, upon NRC approval, will be included in the program.

Collection of data activities will be:

- Collect data from the Northrop TRIGA reactor decommissioning through direct purchase order from the dismantling contractor, Chem-Nuclear Systems, Inc.
- Continue decommissioning data collection for projects already included in the ENFDP program. The ongoing projects include:
 - Data collection from TMI-2 recovery efforts.
 - Data collection, as released by DOE, for the Shippingport decommissioning project.
 - Final data collection from the Humboldt Bay safe storage preparation.

Analysis and reporting activities will include:

- Prepare a project summary report for Humboldt Bay for NRC publication upon completion of preparation for safe storage.
- Prepare summary status reports for NRC publication for TMI-2 and Shippingport.
- Continue summary comparison study for research reactors.
- Develop data from NUREG/CR-2601 on LWR post-accident decommissioning and input into the computer file concurrently with TMI-2 data for eventual comparisons.
- Prepare a project summary report for the Northrop TRIGA reactor when data is available.

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9. Letter, R. L. Miller to Dr. Carl Feldman, "Final Report - International Nuclear Reactor Decommissioning Planning Conference," dated September 14, 1985.
10. Letter, R. L. Miller to Dr. Carl Feldman, "Final Report - International Nuclear Reactor Decommissioning Planning Conference Financial Summary," dated September 30, 1985.

Hydrogen Mitigative and Preventive Schemes Program

(FIN No. A 1336)

Branch Contractor's Annual Report

Summaries of Research
Conducted By
Sandia National Laboratories
During FY 1985

Prepared for
The Chemical Engineering Branch
Division of Engineering Technology
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission

Principal Investigators:
Marshall Berman and Lloyd S. Nelson

This informal document contains information of a preliminary nature and was prepared primarily for interim use in reactor safety programs in the United States. Thus, it is subject to revision or correction, does not constitute a final report, and should not be cited as a reference in publications.

EXECUTIVE SUMMARY

1. HYDROGEN MITIGATIVE AND PREVENTIVE SCHEMES

The objective of the Hydrogen Mitigative and Preventive Schemes program is to provide the NRC with information to evaluate proposed equipment concepts and operational schemes to prevent or mitigate the effects of hydrogen combustion during hypothetical LWR accidents. To provide this information, we are investigating the operability and consequences of operation of deliberate ignition systems and their components during hypothetical hydrogen-producing accidents in nuclear power plants.

We have demonstrated that the damaging aspects of hydrogen combustion may be heightened substantially by the presence of a metallic iron aerosol during the burn. Two replicate experiments were performed in the 5.1 m³ VGES vertical combustion tank with 6.5 v/o hydrogen/air mixtures. In each experiment, a 2-s long burst of gas dispersed 1 kg of sponge iron powder (76 percent through 44 micron screen) throughout the chamber gases. (The metal simulated a reduced aerosol related to fuel and structural materials generated during a core degradation accident.) One second after dispersal, in each experiment, the gas mixture was ignited with a bridge-wire. Vigorous deflagrations occurred in both experiments as indicated by pressure and temperature sensors located throughout the chamber. Both reddish brown and black debris were found after the burns, suggesting the formation of Fe₂O₃ and Fe₃O₄, respectively.

The peak pressure rise produced when the metallic aerosol was present was ~0.3 MPa, compared to ~0.1 MPa in control experiments performed identically without the aerosol. Thermocouples exposed to the burning mixtures registered maximum absolute temperatures 50 to 90 percent higher than in the control experiment. Moreover, 95 percent of the hydrogen burned with the aerosol present compared to only about 60 percent without the aerosol. Clearly, the iron aerosol burned rapidly along with the hydrogen, adding its enthalpy of combustion to that of the gas mixture.

Since the 6.5 percent hydrogen/air mixture is normally considered a "benign" composition for deliberate ignition to remove hydrogen from containment--rich enough to burn gently, yet lean enough to produce low pressure and temperature rises--its ability to ignite the metallic aerosol so effectively was unexpected.

In order to learn more about the combustion of the hybrid mixture Fe-H₂-air, we performed further experiments--analogous to those in the 5.1 m³ VGES chamber--in the 0.18 m³ chamber at McGill University. In these tests

EXECUTIVE SUMMARY

43 shots were fired: 9 shots for the first series with quiescent H₂-air mixtures, 11 shots for the second series with turbulent H₂-air mixtures, 12 shots in the third series with only iron powder as fuel, and 11 shots for the fourth series with both H₂ and iron powder. The parameters measured were peak overpressure $\Delta p_M/p_0$ and the rate of pressure rise $(dp/dt)_{\max}$. When the iron aerosol was present, these parameters were usually increased above those recorded in similar experiments without the iron powder.

Also, we have studied the flammability limits of the hybrid mixtures and have found an approximately linear burn-no burn relationship between the concentration of iron aerosol and the amount of hydrogen present in the hybrid mixture. This linear relationship extends between approximately 4 percent hydrogen in air with no iron, the lean limit of hydrogen combustion, to 275 g/m³ of iron aerosol with no hydrogen in the 5.1 m³ VGES chamber; a less linear relationship terminating at 400 g/m³ is observed in a smaller, 0.18 m³ chamber. The importance of scale on the flammabilities of hybrid metal aerosol-gaseous fuel mixtures is demonstrated conclusively.

On the basis of the large- and small-scale experiments, we draw these tentative conclusions:

1. There is synergism between the gaseous and metallic fuels. For example, the metallic aerosol will ignite at significantly lower concentrations with small amounts of hydrogen than with no gaseous fuel present. Also, the hydrogen burns more completely with the aerosol than without.
2. There is apparently a strong effect of the scale of the experiment on the violence of the explosion, with larger peak pressures produced in the larger apparatus for similar mixtures.
3. There is a strong dependence on metal particle size and morphology.

We are studying the effects produced by operating the containment water-spray system while the deliberate ignition system is activated. We have completed an experimental study of resistance-heated igniters exposed to water sprays and gas flows. We have compared glow plug igniters operated at both 12 and 14 V ac (RMS) and determined that the igniters would fail to ignite lean hydrogen-air mixtures when exposed to dry airflows of approximately 7 and 13 m/s, respectively.

EXECUTIVE SUMMARY

Airflows of this magnitude have been shown by CONCHAS-SPRAY calculations to be possible under certain containment conditions during the operation of the water spray system.

We have continued our studies of the effects of liquid water dispersed as droplets on the combustion of hydrogen-air mixtures. We are using the 5.6 m³ FITS chamber for these studies. A new gas dosing system was installed that produces mixtures of hydrogen in air in the FITS chamber with analyzed compositions reproducible to better than 2 percent. Using this system, we have completed a sequence of experiments in which 6.5 and 10 percent mixtures of hydrogen in air were burned in the presence of approximately 100 g/m³ of both 100 and 200 μ m-diameter water drops. We find that compared to control burns performed without water drops, there is little change in the pressure-time relationships for the 10 percent mixture, which lies above the downward propagation limit. In burns at 6.5 percent hydrogen in air, however--below the downward propagation limit--the pressure-time records are altered considerably: peak pressures approximately double, while the reproducibility of the burns increases strongly. We have concluded that the presence of heterogeneous water dispersed in a lean hydrogen-air mixture below the downward propagation limit can considerably enhance the vigor of the burn, probably due to microturbulence produced around the individual droplets.

We have demonstrated the feasibility of using a nonpowered platinum catalytic igniter for the ignition of lean hydrogen-air mixtures, both in a laboratory-scale burner with flowing gases and in the 5.6 m³ FITS chamber with a static mixture. Mixtures as lean as 5.5 percent have been ignited in the laboratory system, while mixtures of both 6.5 and 10 percent hydrogen in air have been ignited in the large-scale experiment. These tests demonstrate that nonpowered igniters for lean hydrogen-air mixtures are feasible and that they can contribute to the safety of light water reactors by providing a backup for the ac-powered igniters in the event of a station blackout.

1. HYDROGEN MITIGATIVE AND PREVENTIVE SCHEMES

(L.S. Nelson and M. Berman, 6427)

The objective of the Hydrogen Mitigative and Preventive Schemes program is to provide the NRC with information to evaluate proposed equipment concepts and operational schemes to prevent or mitigate the effects of hydrogen combustion during hypothetical LWR accidents. To provide this information, we are investigating the operability and consequences of operation of deliberate ignition systems and their components during hypothetical hydrogen-producing accidents in nuclear power plants.

1.1 Effect of Metallic Aerosols on Hydrogen Combustion

(L. S. Nelson and K. P. Guay, 6427; W. B. Benedick, 1131; W. Einfeld, 6324; G. D. Valdez, 6449; J. H. Lee, R. Knystautas, and M. Gaug, McGill University, Montreal, Canada)

We have continued our studies of the effects of metallic aerosols on the combustion of lean hydrogen-air mixtures. We performed experiments both in the 5.1-m³ VGES chamber at Sandia, Albuquerque, and in the smaller chamber of 0.18-m³ volume at McGill University, Montreal, Canada.

1.1.1 Experiments in the VGES Chamber

(L. S. Nelson, 6427; G. D. Valdez, 6449; W. B. Benedick, 1131)

In the October-December 1984 quarterly report,¹ we described a single combustion experiment in the VGES chamber in which a metallic iron aerosol was dispersed throughout a 6.5 percent hydrogen-air mixture just before a bridgewire igniter was activated. There was a substantial increase in the peak pressure recorded during this burn above that of a control burn without the metal aerosol (Figure 1.66, Reference 1).

The iron aerosol used in this single experiment was generated by the gas burst dispersal of 727 g of fine steel shot, AISI C-1018, obtained from Nuclear Metals, Inc., of Concord, MA. This powder is relatively coarse, as shown in the scanning electron micrograph in Figure 1. Note the smooth, spheroidal form of the material. Particle dimensions are on the order of 100 to 200 μm .

In new experiments performed during February 1985, we repeated the earlier experiment with a similar hydrogen composition, but with a finer iron powder (Ancor 2023 sponge iron powder obtained from the Hoeganaes Corporation of Riverton, NJ). A scanning electron micrograph of this new material is reproduced in Figure 2 at the same magnification as Figure 1. Note the more angular structure of this

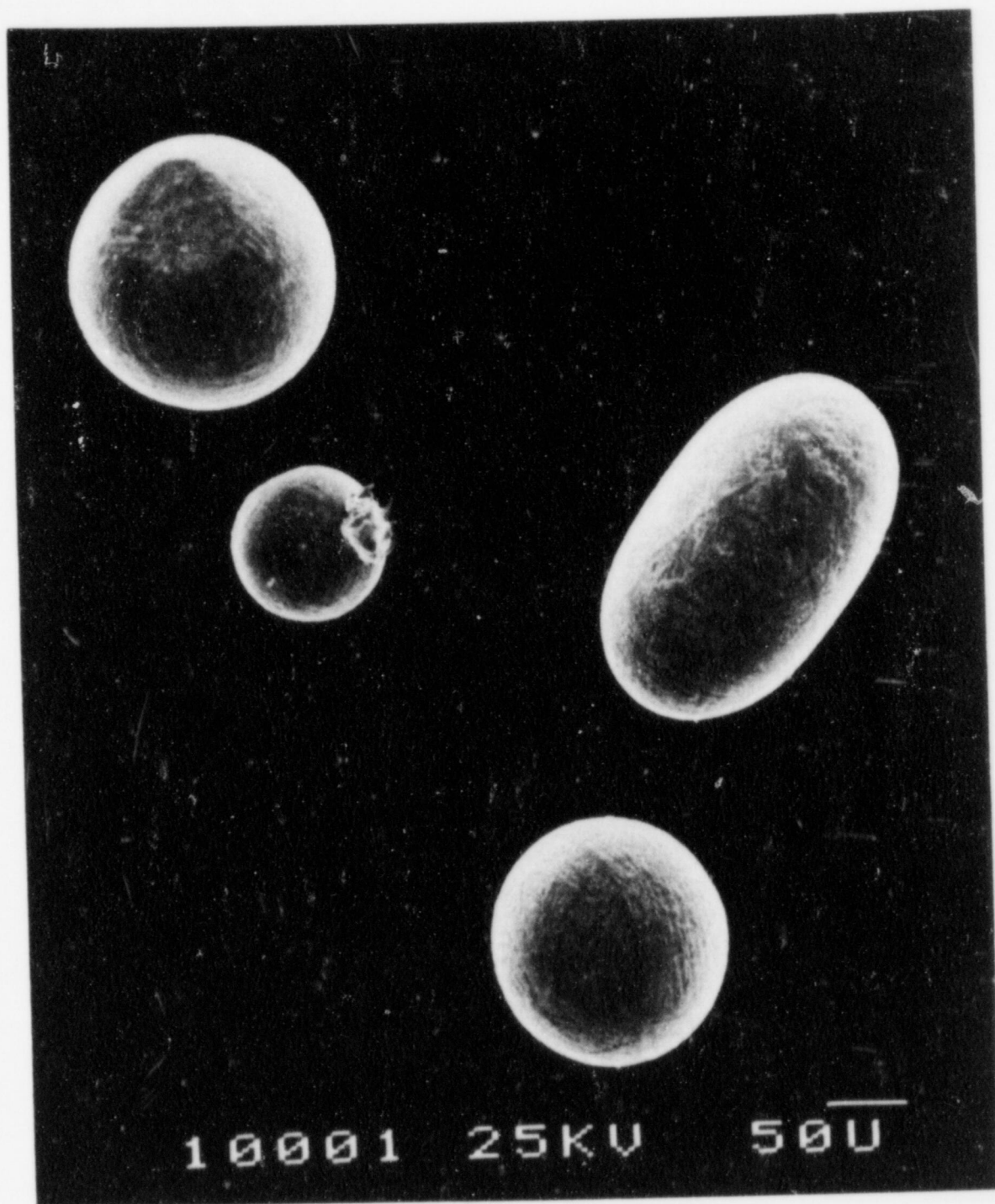


Figure 1. Scanning Electron Micrograph of Iron Powder Used in First Metallic Aerosol/Hydrogen Combustion Experiment in the VGES Chamber. Scale bar is 50 microns long.

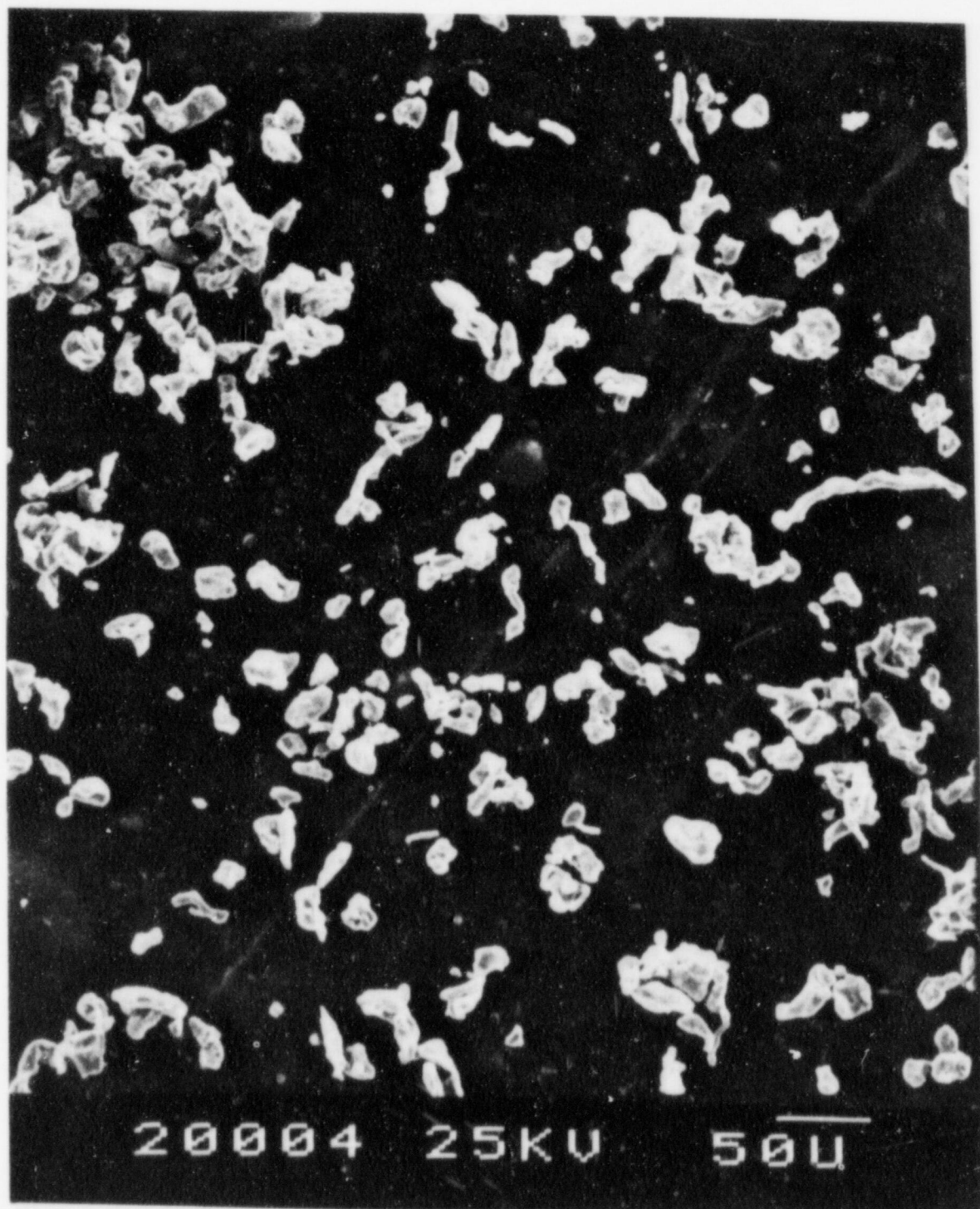


Figure 2. Scanning Electron Micrograph of Iron Powder Used in Second Series of Metallic Aerosol/Hydrogen Combustion Experiments in the VGES Chamber. Scale bar is 50 microns long.

material. Particle dimensions here are on the order of 10 to 40 μm , about an order of magnitude smaller than the material used in the previous experiment.

In the new experiments, we used the bowl-type disperser described in Reference 1 and shown in Figure 1.65 of that reference. This dispersing system is scaled up from the one used in the small-scale experiments described in Reference 1. The experiments in the VGES chamber were performed by dispersing 1 kg of the sponge iron powder throughout the hydrogen-air mixture in the chamber with a 2-s long burst of air; the mixture was ignited 1 s after dispersal with a bridgewire. The combustion diagnostics were those used previously.²

When the iron aerosol was present, vigorous deflagrations occurred, as indicated by pressure and temperature sensors located throughout the chamber. The peak pressure rise recorded when the metallic aerosol was present was ~ 0.3 MPa, compared to a rise of ~ 0.1 MPa in control experiments performed identically without the aerosol. This is shown in Figure 3. Also, thermocouples exposed to the burning mixtures registered maximum absolute temperatures 50 to 90 percent higher than in the control experiment (this is shown in Figure 4 for a thermocouple placed at about the midpoint of the tank). In addition to the enhanced thermocouple response, static calorimeters (brass plates with temperature-sensitive labels) showed a considerable increase in heat flux in the experiments with the aerosol compared to those without.

A further indication of the vigor of the burn in the presence of the iron aerosol was that both reddish brown and black debris was found in the chamber afterward, suggesting the formation of Fe_2O_3 and Fe_3O_4 , respectively. Moreover, 95 percent of the hydrogen burned with the aerosol present compared to only about 60 percent without the aerosol. Clearly, the iron aerosol burned rapidly along with the hydrogen, adding its enthalpy of combustion to that of the hydrogen-air mixture.

In order to compare this work with other hydrogen burns, we show peak pressure rises in Figure 5 for the two experiments performed. These data points have been superimposed on the figure taken from Reference 2 which shows the results of previous combustion experiments performed both in the VGES chamber and in vessels in other laboratories, as indicated in the code in the lower right of the figure. It should be noted that the peak pressure obtained here is several times that produced by the hydrogen alone, and far exceeds the calculation for the adiabatic, constant volume, complete combustion burns (the solid line in Figure 5).

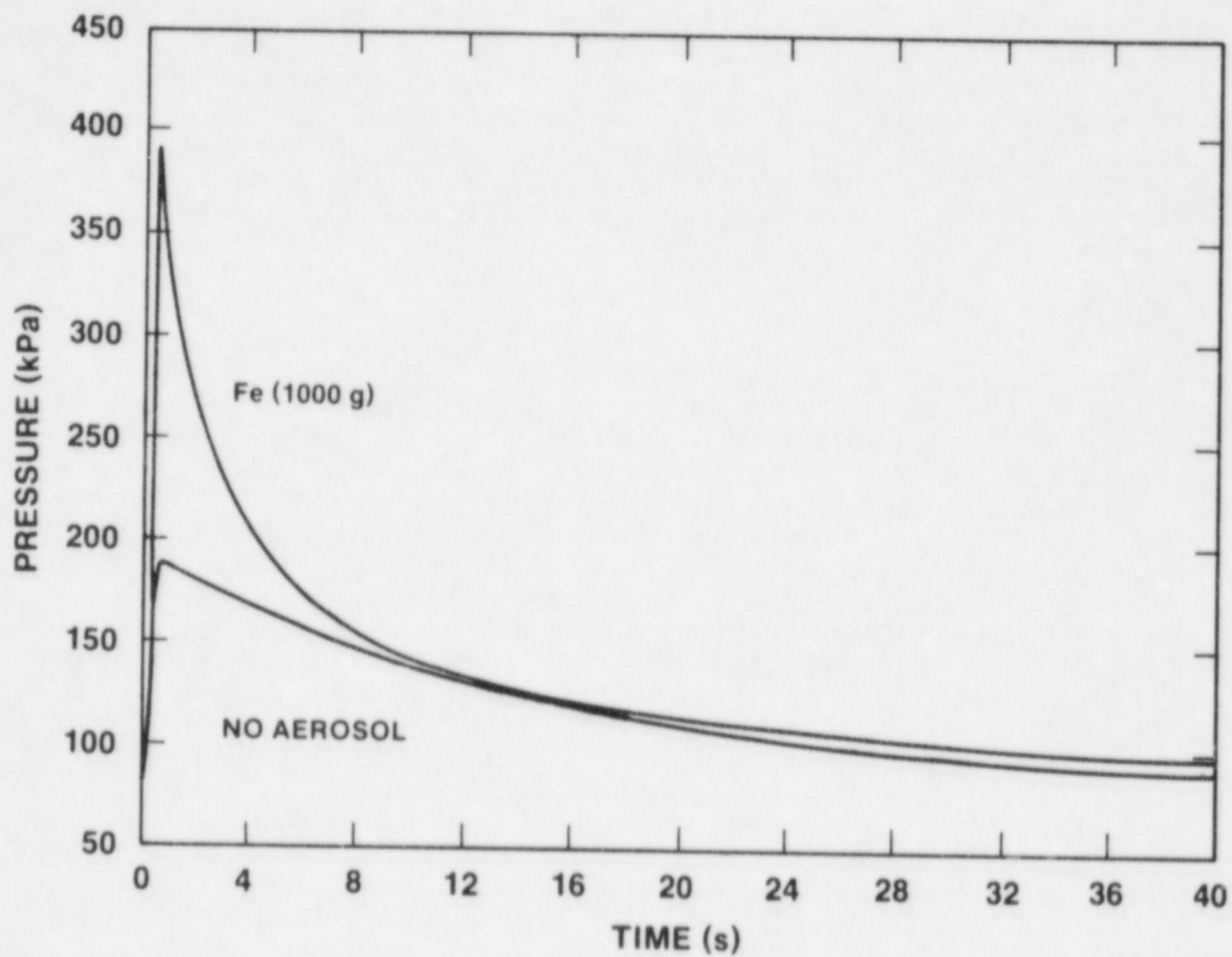


Figure 3. Pressure Records for Iron Aerosol/Hydrogen Combustion Experiments Performed in the VGES Chamber. Upper curve was recorded with the metallic iron; lower curve was recorded without the iron. The hydrogen concentration in both experiments was 6.5 v/o.

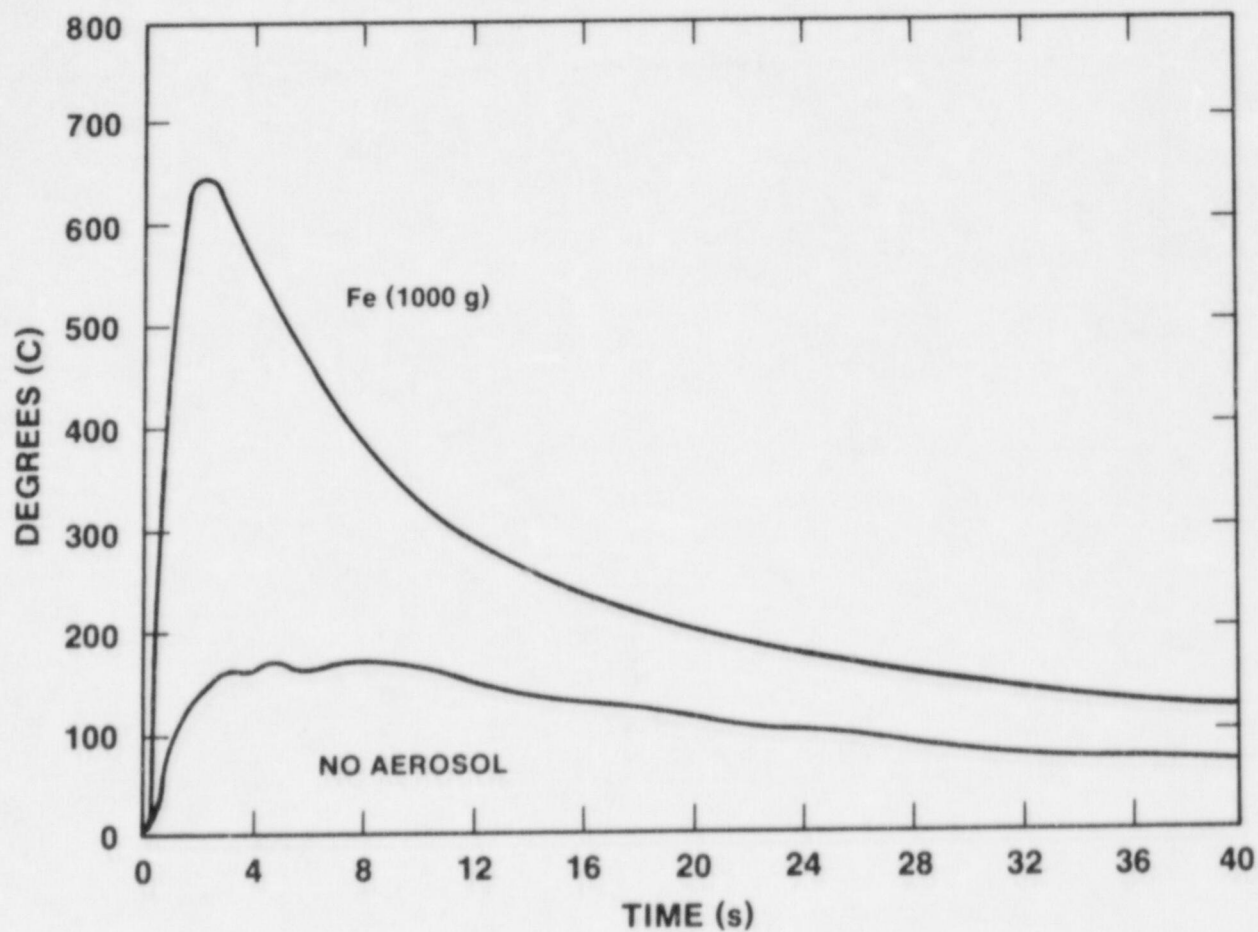


Figure 4. Typical Thermocouple Traces for Iron Aerosol/Hydrogen Combustion Experiments Performed in the VCES Chamber. Upper curve was recorded with the metallic iron; lower curve was recorded without the iron.

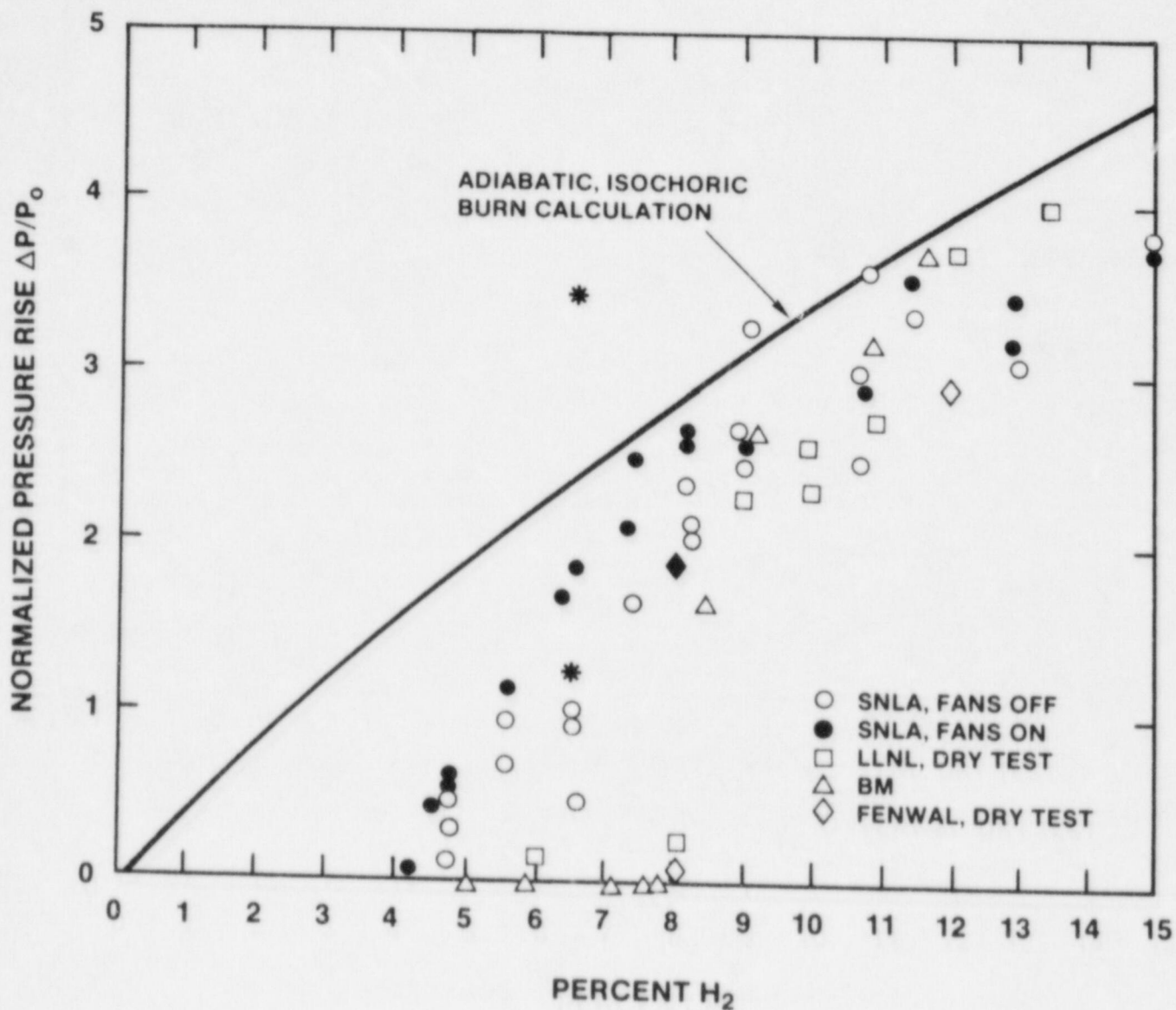


Figure 5. Normalized Pressure Rises Produced With (Upper Asterisk) and Without (Lower Asterisk) Iron Aerosol in a 6.5 Percent Hydrogen-Air Burn in the VGES Chamber Compared With Earlier Experiments. For original plot and explanation of laboratory code, see Reference 2.

Since the 6.5 percent hydrogen/air mixture is normally considered a "benign" composition for deliberate ignition to remove hydrogen from the containment of a nuclear plant--rich enough to burn gently, yet lean enough to produce low pressure and temperature rises--its ability to ignite the metallic aerosol so effectively was unexpected. (In terms used in other fields of combustion, we have produced a "hybrid dust explosion," a mixture of gaseous fuel and a combustible dust (aerosol). Analogs of considerable importance are coal dust-methane mixtures, a persistent hazard in coal mines.)

1.1.2 Laboratory Scale Experiments: Flammability of Lean Hydrogen-Air Mixtures Seeded With the Fine Iron Aerosol
(R. R. Knystautas, G. H. Lee, and M. Gaug, McGill University)

Preliminary experiments in the VGES tank have indicated that the presence of dispersed fine iron aerosol in a very lean H₂-air mixture (6.5 percent H₂-air) enhances substantially the peak overpressures produced when the hybrid mixture of H₂-air-iron is ignited. These observations have naturally created some concern and aroused interest in the combustion behavior of such hybrid mixtures over a broader range of parametric conditions. In response to this and in direct support of the VGES experiments, the 0.18 m³ explosion vessel used in general dust combustion studies at McGill University was activated to specifically address this need. Since the McGill facility, once fully operational, can be run at a rate of up to one shot per hour, it was felt that a broad range of parametric conditions could be explored very rapidly to assist in planning further large-scale VGES experiments. The McGill facility can simulate rather well but on a smaller scale the main thrust of the VGES experiments.

In late March 1985, appropriate alterations were made to the McGill 0.18 m³ dust explosion vessel to conform to the VGES test. Detailed experiments were begun on March 31, 1985. The McGill experiments were performed in four stages to provide broader base line data to enhance the capability of data analysis for the H₂-air-iron flammability tests. We proceeded in four phases. The first phase repeated the quiescent H₂-air burns in our chamber in the range of 6.5 percent H₂ to the lower flammability limit of hydrogen in air (~4 percent). The data on $\Delta p_M/p_0$ as well as the rate of pressure rise $d\Delta p/dt$ were compared with existing data obtained previously in the VGES tank by Benedick et al.² This served as a reference to assess the characteristics of our vessel (e.g., heat losses). The second phase repeated the turbulent burn series, again with H₂-air

only. The turbulence was generated by the same air jet system that was used to disperse the iron powder in later experiments. The third series of experiments was performed with iron powder only, to determine the pressure rise and flammability limits with just the iron as fuel. The fourth phase was the hybrid H₂-iron-air series. Starting with the 6.5 percent H₂ and 100 g/m³ iron, we proceeded to the limit (i.e., ~4 percent H₂) at intervals of 0.5 percent in H₂. A 50-g/m³ iron concentration at different H₂ concentrations also was carried out for comparison.

1.1.2.1 Experimental Details

The McGill University dust explosion vessel has a volume of approximately 0.18 m³ and is cylindrical in shape with rounded dome-shaped ends. The cylinder itself consists of a straight portion (48.2 cm in diameter x 54.2 cm in length) capped by two dome-shaped ends which are each 48.2 cm in diameter with a mean equivalent cylindrical length of approximately 22 cm each. Thus one can think of the vessel as an equivalent cylinder of (48.2 cm in diameter x 98.2 cm in length). The explosion vessel is provided with a large number of sealable ports for a variety of inlets and outlets as required. A schematic diagram of the apparatus is shown in Figure 1.67, Reference 1.

In a given experiment, the mixture in the explosion vessel is prepared by manometric measurements of partial pressures. The percentage hydrogen composition is precalculated and the partial pressure determined. The air in the vessel is then partially evacuated to below the day's barometric pressure and carefully adjusted at the precalculated subatmospheric pressure. Hydrogen is then added up to the full barometric pressure. The contents are then recirculated for 30 min with a Bellows-type pump (Model MB-158) to achieve a well-mixed H₂-air mixture. The pump intake is connected to the top of the explosion vessel via a rockwool filter and is force-exhausted via interior tubing to the very bottom loop. If the experiment involves iron aerosol, then the appropriate amount is preweighed, placed in a stainless steel circular cup, and inserted at the very bottom of the explosion vessel. (The metal used here is Ancor 2023 sponge iron powder, the same as that used in the VGES experiments described in the previous section.) The dispersion is by means of an impinging 12-mm diameter tube which is connected to a 1 L vessel which is pressurized with the same H₂-air mixture as the mixture in the explosion vessel. Hence the same composition H₂-air mixture (or air in iron-air flammability tests) is used to disperse the iron aerosol. The pressure of the dispersal gas is set at 214.7 psia. The dispersal gas is injected into the main vessel by means of an electrically activated solenoid valve. After dispersal

the mixture is ignited by a strand of exploding magnesium fibers (extracted from blue-dot photo flash bulbs). The strand is twisted into a cylindrical shape typically 4 mm in diameter and 2 cm long and clamped between two alligator clips in the center of the explosion vessel. The ignition is achieved by shorting out 110 V ac with a delay-activated relay switch. Diagnostics involve pressure measurement in the explosion vessel using a PCB 113A24 piezoelectric transducer. The calibration of the particular transducer used is 5.14 mV/psi (SN 3060/3304).

1.1.2.2 Results

We fired a total of 43 shots; 9 shots for the first series with quiescent H₂-air mixtures, 11 shots for the second series with turbulent H₂-air mixtures, 12 shots in the third series with only iron powder as fuel, and 11 shots for the fourth series with both H₂ and iron powder. The results for the peak overpressure $\Delta p_M/p_0$ and the rate of pressure rise $(dp/dt)_{\max}$ were recorded for all the shots fired. The peak overpressure results are plotted in Figure 6 together with the previous data of Benedick et al.² obtained from the VGES tank. An expanded scale exposition of the McGill data alone is shown in Figure 7 for the sake of clarity. The $\Delta p_M/p_0$ for the first series where the H₂-air mixture is quiescent are in accord with the previous VGES data. They are slightly lower because in the present experiment ignition is at the center of the vessel while for the VGES data, ignition is near the bottom of the tank. Thus, buoyancy effects will be more prominent in the present experiment. The turbulent data of the second series agrees well with the VGES data. With a higher burning velocity, buoyancy plays a lesser role. With an iron concentration of 100 g/m³, a significant increase in the overpressure is observed for 6.5 percent H₂. Specifically with 6.5 percent H₂, $\Delta p_M/p_0$ is 1.45, while the addition of 100 g/m³ of iron results in an increase of $\Delta p_M/p_0$ to a value of 2.534. This result is in accord with that reported for the VGES experiments where an increase in $\Delta p_M/p_0$ by about a factor of two is observed. Thus we confirm the observation that the addition of a small amount of iron aerosol of the order of 100 g/m³ to a 6.5 percent H₂-air mixture gives rise to about a two-fold increase in the peak overpressure developed. The peak rate of pressure rise which gives an indication of the burning velocity increases also by a factor of two when iron is added (100 g/m³) to a mixture of 6.5 percent H₂. For example, for 6.5 percent H₂ under a turbulent condition, $(dp/dt)_{\max}$ is 643.5 kPa/s, an increase by a factor of two. This could be due to the increase in the flame temperature as a result of the added fuel. However, the percentage increase in the peak overpressure due to iron addition decreases rapidly when the concentration of H₂ decreases. For example, at 4.5 percent

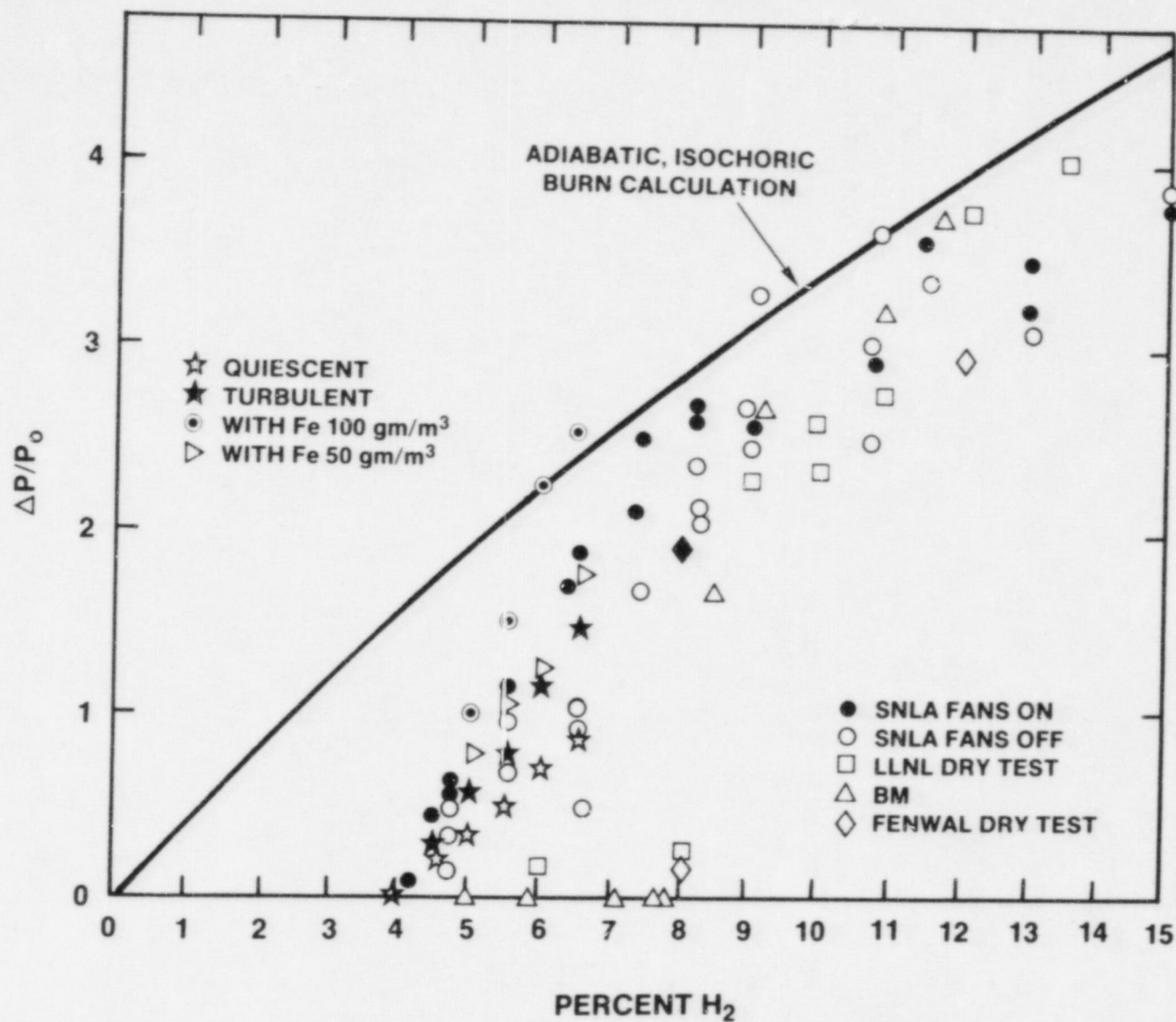


Figure 6. Comparison of McGill Combustion Experiments With Iron Aerosol in Lean Hydrogen-Air Burns With Earlier Experiments. Data are superimposed on figure taken from Reference 2.

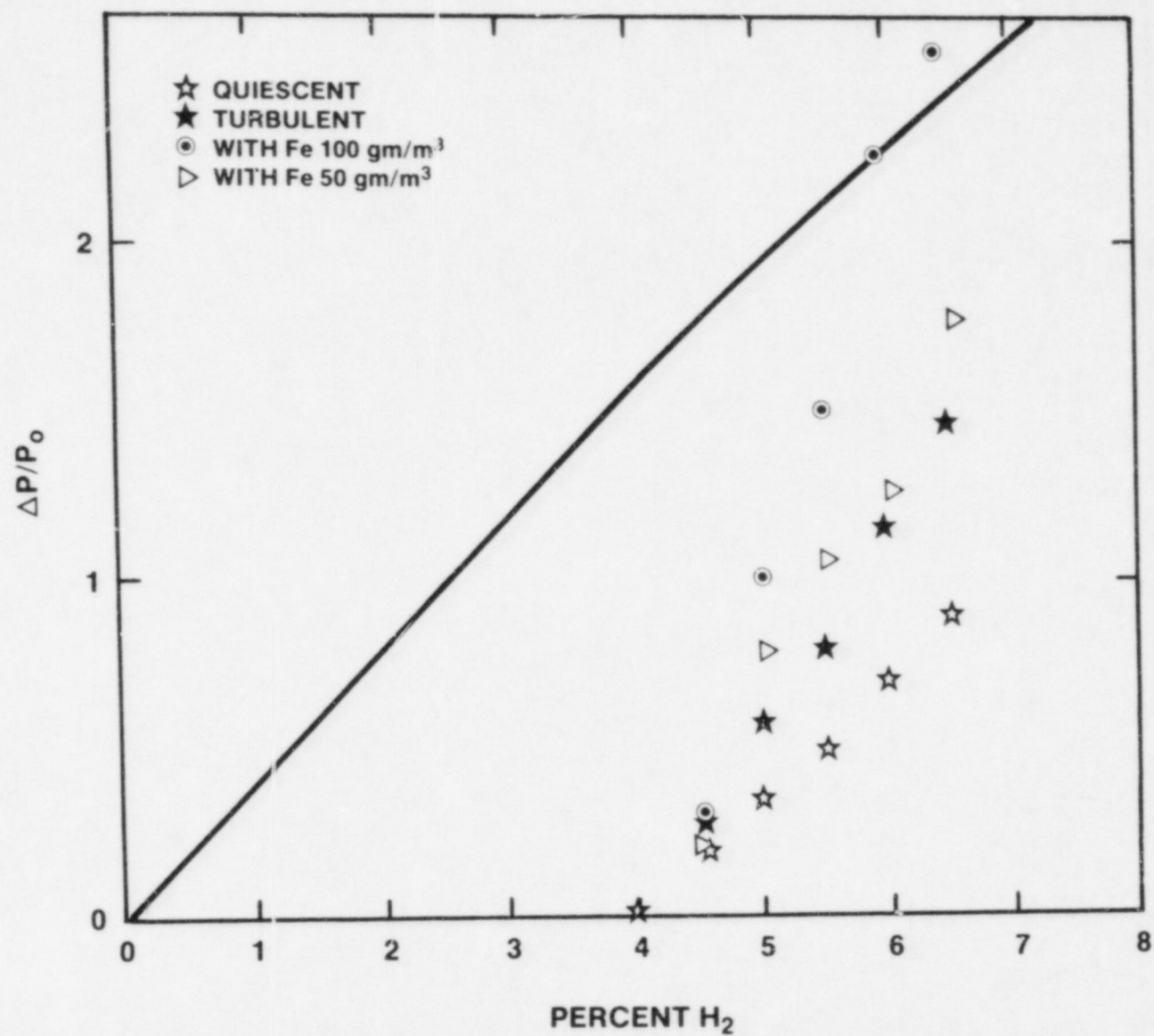


Figure 7. McGill Data from Figure 6 Shown Separately for Clarity

H₂, the $\Delta p_M/p_0$ is 0.32 and is practically identical to the value of $\Delta p_M/p_0 = 0.322$ for the case of no iron aerosol at the same 4.5 percent H₂ concentration. Compatible with this result, we also do not observe any change in the lean limit with the addition of iron aerosol, i.e., we cannot ignite 4 percent H₂ under the same turbulent condition both with and without the addition of iron. However, this result is expected to be different if the iron aerosol particle size is decreased. The particle size of the iron powder used in the present experiment is about 40 μm . Experiments with other aerosols less than 10 μm have indicated that the behavior is almost identical to that of a homogeneous mixture. Thus it may be possible to have a more pronounced effect at the very lean concentration around 4 percent H₂ if the iron aerosol particle size is smaller than 40 μm . The flammability limit may also be altered according to the Le Chatelier law in this case. The experiments with iron powder only indicate that the lean limit for iron is between 300 and 400 g/m³ in the present apparatus. No attempts have been made in this series of experiments to determine the precise value of the lean limit itself. However, it is certain that the concentration of 100 g/m³ of iron alone cannot sustain a flame.

We may conclude that the addition of about 100 g/m³ of iron results in an almost two-fold increase in the peak overpressure (hence temperature) in a mixture of 6.5 percent H₂ which is usually considered "benign." Although this effect is greatly diminished and no extension of the lean limit was observed in the present experiment, particle size is expected to play an important role. Experiments should be carried out with small particle size before definitive conclusions can be drawn regarding the application of Le Chatelier's rule to extend the lean limit.

1.1.3 Flammability Limits of Hybrid Hydrogen-Iron-Air Mixtures

(L. S. Nelson and K. P. Guay, 6427; G. D. Valdez, 6449; J. H. Lee, R. Knystautas, and M. Gaug, McGill University, Montreal, Canada)

The ease of ignition of the iron aerosol clouds in the presence of a small amount of hydrogen described in References 1 and 3 prompted us to examine the limits at which combustion will occur in this hybrid mixture of gaseous and solid fuels in the presence of air. (Burns such as these are also called hybrid dust explosions, for example, coal dust-methane mixtures⁴ and mixtures of polyvinyl chloride dust with methane or propane.⁵)

The rule of Le Chatelier is often used to determine flammability limits of homogeneous gas mixtures.⁶ The rule

predicts that for a mixture of two fuels, there should be a linear relationship between the flammability limits and the concentrations of each component. Although this rule has been used in studies of coal dust*-methane-air mixtures in small-scale apparatus, its applicability to metal aerosol*-gaseous fuel-oxidizer mixtures has not been examined. We have used the rule here to correlate flammability limits in the iron aerosol-hydrogen-air system determined in both the VGES and the McGill experimental chambers. The sponge iron powder described in Reference 3 was used in both chambers.

In this work, attention was paid also to the importance of scale on the aerosol combustion. Apparently, aerosol flame thicknesses are large, often as large as 1 m. Thus, the dimensions of the chamber in which the combustion is performed must be at least on this order to give valid information of flammability limits. The VGES chamber is large enough to eliminate the adverse effects of small scale. However, due to the difficulties and costs involved with running tests in the large vessel, many tests were also performed in the small McGill chamber.

In earlier work, the lean flammability limit of hydrogen-air mixtures has been determined to be about 4 percent hydrogen in air in both the large² and small chambers. We then determined the flammability limit of the iron aerosol alone in air.** In the smaller chamber, it was found to be on the order of 400 g/m³, roughly four times that reported in the literature.⁸ No information, however, is given in the literature about the details of vessel size or configuration nor the particle-size range of the iron aerosol. The analogous lean limit for iron aerosol in air, determined in the VGES chamber, was somewhat lower, namely 275 g/m³. It is felt that the larger dimensions of the chamber are responsible for this lower limit.

We then determined go-no-go limits for combustion in the two chambers as we proceeded from the gaseous fuel to the solid fuel; we found considerable deviations from the linear relationship predicted by the Le Chatelier rule. This deviation for the two chambers is shown in Figure 8. (The data are normalized at both lean limits to the particular flammability limits determined in that chamber.) It will be noted in

* In this report, a dust and an aerosol are considered to be identical.

** Aerosol concentrations were estimated by assuming all metal powder was dispersed quantitatively and uniformly throughout the chambers.

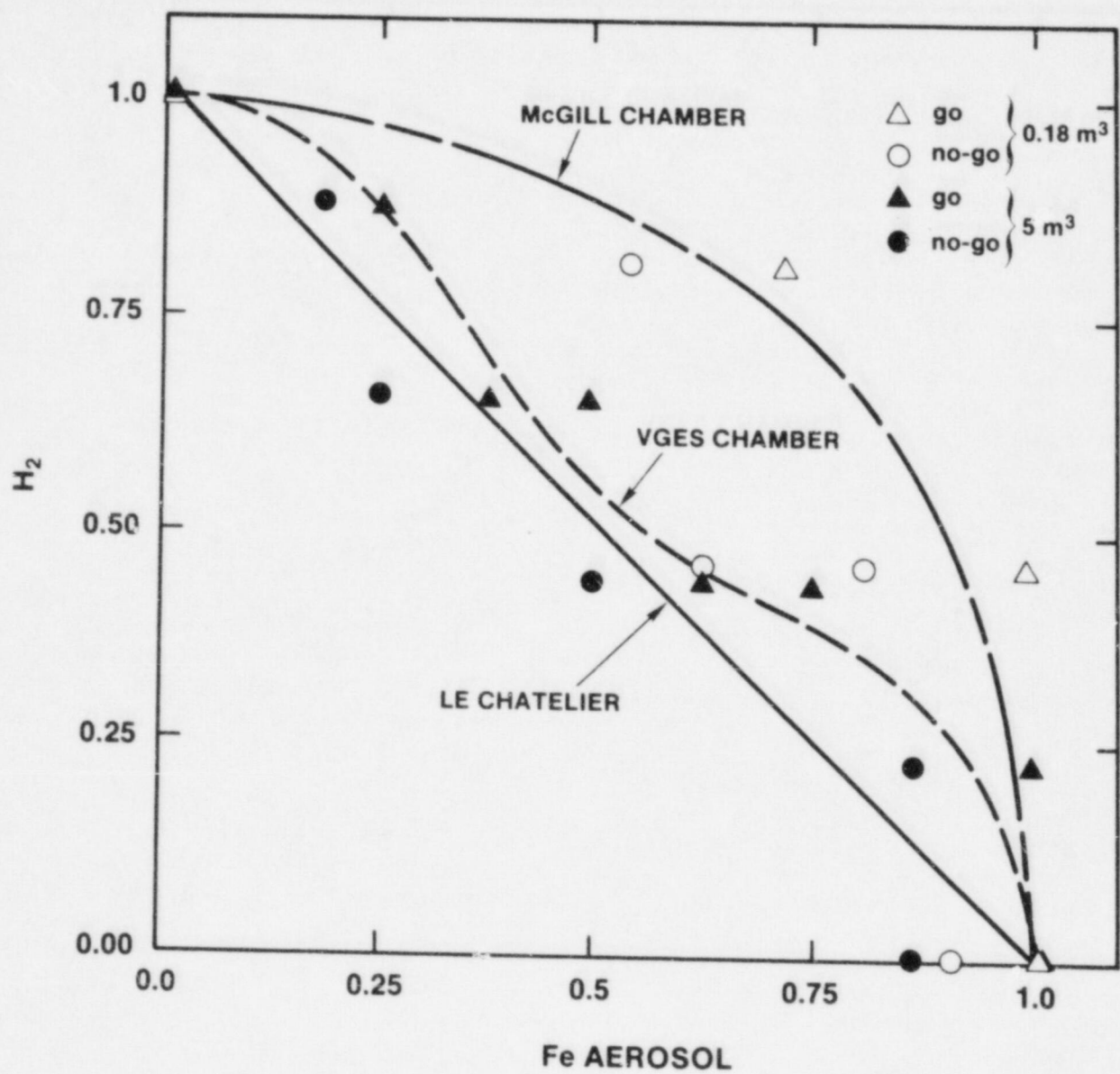


Figure 8. Flammability Limits of H_2 -Fe Aerosol-Air Hybrid Mixtures for Large and Small Scale

Figure 8 that although the general trend of flammability follows the Le Chatelier rule, the plot shows considerable deviation at the pure fuel extremes for the large chamber and over the entire range for the smaller chamber.

We conclude from Figure 8 that Le Chatelier's rule does not apply well to our hybrid metallic iron aerosol-hydrogen mixtures. This suggests that the hybrid lean flammability limits are not dependent on the energetics of the combustion, upon which the Le Chatelier rule is based, but instead are governed largely by the fluid dynamics of the particular combustion geometry used. Even though the Le Chatelier rule is not valid for the hybrid mixtures studied, however, it may provide an approximate base line from which to study other hybrid mixtures.

It can also be seen in Figure 8 that the results obtained in small-scale apparatus may be misleading when the flame thicknesses are larger than the vessel dimensions. We recommend that a large-scale apparatus be used whenever possible when studying lean flammability limits of either aerosols (dust) alone or hybrid aerosol-gas mixtures.

1.2 Behavior of Hydrogen Igniters During the Operation of Water Sprays in Containment

(L. S. Nelson, K. P. Guay, and P. A. Snowball, 6427)

During the operation of water sprays in containment, airflows with velocities in the range 5 to 10 m/s may be generated by the entrainment of air caused by the downward falling drops.⁹ Earlier we reported¹⁰ the behavior of resistively heated hydrogen igniters exposed to various airflows. In Figure 1.2-3 of Reference 10 we presented a plot of surface temperature as a function of dry airflow for two igniters, the Tayco helical igniter operated at 120 V ac (RMS) and the General Motors AC 7G diesel glow plug operated at 14 V ac (RMS).

We recently learned that some ice condenser plants operate their glow plug igniters at 12 V ac. For completeness, then, we set up our glow plug igniter again as described in Reference 10. The igniter had four Type K chromel-alumel thermocouples spot-welded to its surface in the positions shown in Figure 9. The outputs of these thermocouples, properly referenced thermally, were recorded as a function of time by the DAASY III data acquisition system. Airflows up to about 25 m/s were generated by either an industrial vacuum cleaner or a squirrel cage blower. To vary airflows, the vacuum cleaner was exhausted through tubes of different diameter. Different airflows were obtained with the blower

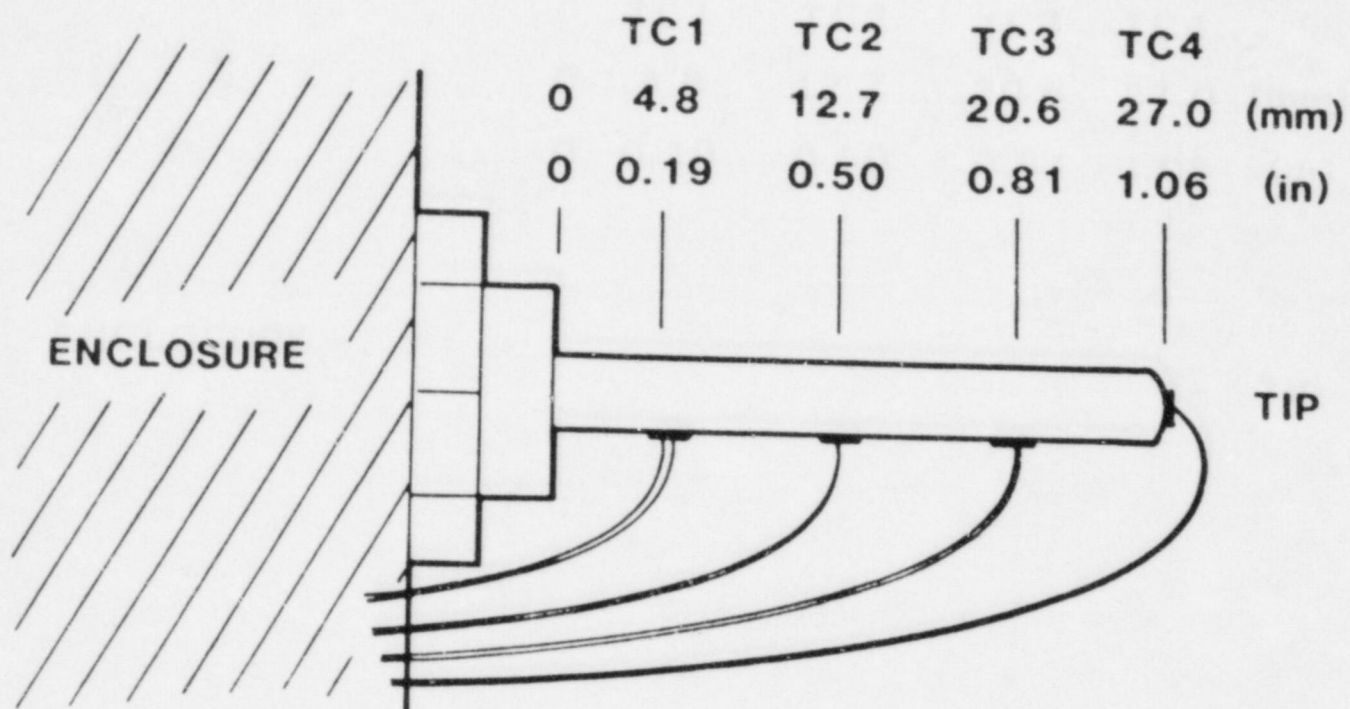


Figure 9. Diagram of the Placement of the Four Thermocouples on the General Motors AC-7G Cylindrical Glow Plug Igniter When Used in the Exposure Tests at Various Spray Fluxes and Air Flows

by placing the igniter at various distances from the outlet. Flow velocity in both cases was measured with an MKS baratron gauge operated as a pitot tube.

A new plot of surface temperatures of the glow plug operated at both 12 and 14 V ac (RMS), measured at the terminals of the glow plug, as a function of airflow is shown in Figure 10. The dashed line labeled IGNC indicates the minimum temperature at which the glow plug ignited a 6.5 percent hydrogen-air mixture in earlier tests,¹¹ namely, 721°C (1330°F). Note that the surface temperature of the glow plug falls below this temperature at airflows of ~7 and ~13 m/s for operation at 12 and 14 V ac (RMS), respectively. The latter velocity is somewhat higher than the value of ~10 m/s measured in our earlier experiments.¹⁰

1.3 Air Currents Driven by Sprays in Reactor Containment Buildings (K. D. Marx, 8363)

Water spray systems in nuclear power plant containments may be actuated during the time in which hydrogen igniters are required to function. A question of current interest in the field of hydrogen control in reactors is whether the sprays can affect the performance of the igniters. In spray tests in the Zion containment building, air currents were observed on the floor that were later conjectured to be of the order of 10 m/s. This is of concern for the performance of thermal igniters located in the upper compartment of ice condenser containments, because air velocities of this magnitude, if present, may be capable of reducing igniter effectiveness in either of the following ways: (1) they may blow spray onto the igniters from the side, or from below, hence nullifying the effect of the overhead shields, or (2) they may provide convective cooling sufficient to reduce the temperature of the igniters below the ignition point.

In FY85, we performed a large number of two-dimensional numerical simulations of the motion of the water spray droplets coupled with the flow of air in the upper compartment of containment buildings. These simulations have been carried out with the CONCHAS:SPRAY computer code. The geometry and spray flux which have been used for the calculations are those specified for the Sequoyah and Catawba nuclear plants.

The calculations have achieved the following three primary objectives:

1. The observation of air currents near the floor of the Zion containment building during water spray tests appears to be substantiated.

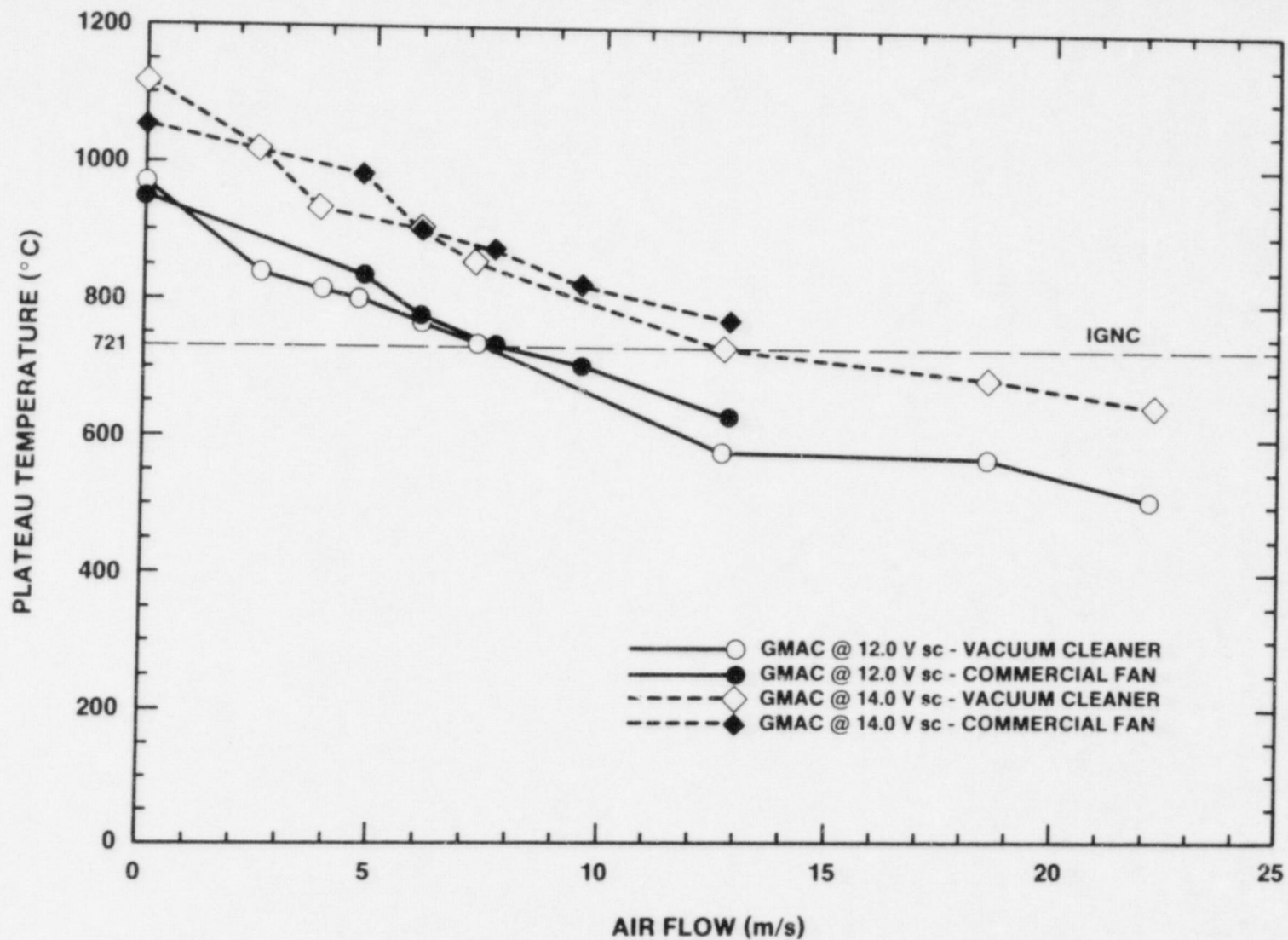


Figure 10. Surface Temperatures of a General Motors AC-7G Glow Plug Exposed to Dry Airflows at Various Velocities. Airflows were produced by a squirrel cage blower (closed symbols) and the exhaust from an industrial vacuum cleaner (open symbols). The dashed line corresponds to the minimum temperature at which a glow plug ignited a 6.5% hydrogen in-air mixture.

Our results confirm that radial (horizontal) flows of 10 to 12 m/s might occur in a containment building shell, i.e., in a containment building lacking any intervening internal structures. In this case, the air flow pattern strongly resembles one which is a solution of the equations for steady-state incompressible inviscid flow. This flow structure is that of a confined vortex ring. The flow velocities are such that the spray is swept toward the center of the building, falls toward the floor, and then tends to be blown radially outward near the floor. The large spray droplets hit the floor, while the smaller ones may circulate back up.

2. The sensitivity of the air flow patterns and velocities to various spray parameters has been examined. These include spray flux, spray ring location, droplet size, and spray injection characteristics.

Theoretical models have been derived which provide insight into the structure of the air currents and which permit order-of-magnitude estimates of the velocities. The theory predicts a weak dependence of flow velocity on spray flux which is observed in the computational results.

The distribution of droplet sizes has a direct bearing on droplet trajectories, and it therefore affects the magnitude of the air flow velocity, although not dramatically. Not surprisingly, the location of spray rings (uniformly spaced or concentrated) has a very significant influence on the flow pattern. Hence, it can affect the flow velocity in certain locations.

It has been established that the modeling of droplet coalescence is important for a correct description of the interaction between the spray and the air currents. To this end, a computational technique which describes this collisional process has been developed and implemented. Coalescence reduces the population of small droplets and increases the size of the large ones. Among other things, this has the effect of reducing the amount of water that can circulate upwards, because only the lighter droplets avoid falling out at the floor of the building.

It has been shown that the peak air flow velocities are very sensitive to the locations of the spray rings relative to the structures in the containment building.

The computational results are relatively insensitive to the viscous stresses imposed on the flow. This is particularly important because of the fact that the

Reynolds stresses are simulated numerically by a Sub-grid Scale (SGS) model, and its efficacy for this particular problem is not known.

3. Some of the effects of structures inside the containment building and of air fans have been computed.

Structures that have been considered are ice condensers and steam generator doghouses. As expected, when the domain inside the building is blocked by first the ice condensers and then the steam generators, the flow velocities are successively diminished. However, the flow is very sensitive to the location of structures insofar as this influences the droplet fall distance and flow constrictions. This means that the presence of structures may not reduce the flow velocity in localized regions.

The air fans have a rather small influence when averaged around the azimuthal direction. However, their effect could be strong in localized regions.

The geometry is assumed to be axisymmetric in these calculations. Hence, some caution should be exercised in their interpretation. It is conceivable that constriction of the flow between (three-dimensional) structures in a real reactor could lead to local peaks in velocity which exceed the predicted peak velocities. This is particularly true near the ducts and gratings which provide for the air-fan exhaust. Except for this caveat regarding three-dimensional effects, the estimates of air flow velocities should be conservatively high. The primary reason for this is that the calculations have used very regular geometries. This permits the formation of large, strong, well-ordered vortices, which lead to greater flow velocities. From this standpoint, the two-dimensionality is a conservative assumption. The inclusion of more structures in the building would impede the air flow. Another less significant conservative factor is that the eddy viscosity has been purposely reduced.

The calculations are essentially isothermal and are carried out under the assumption that the water droplets exist in an atmosphere of saturated air. Hence, the effects of droplet evaporation and thermal convection driven by reactor core heating have not been accounted for.

Experimental data indicates that the igniters will fail due to convective cooling if they are exposed to air flows in excess of ~ 10 m/s. Our results indicate that there are regions of significant extent where the flow velocities are

less than this. (The peak velocities are 9 and 14 m/s in the calculations which we deem most realistic.) This appears to be an optimistic indication for igniter survival.

We have not been able to offer quantitative answers to the question of igniter operability due to sprays blowing up under the igniter shields and onto the igniters. The spray droplet statistics in these calculations are not good enough to provide reliable estimates of upward spray fluxes, as these fluxes are crucially dependent on the population of small droplets. Before a definitive calculation of spray flux could be undertaken, the number of droplet parcels would have to be increased, and it would be necessary to thoroughly assess the accuracy of the coalescence model.

1.4 Combustion of Hydrogen in the Presence of Suspended Water; Monodisperse Drops (L. S. Nelson and K. P. Guay, 6427)

In an earlier report¹ we described the performance of a pair of rotating disk water drop generators installed in the FITS chamber. We showed that it was possible to achieve concentrations of suspended water on the order of 100 g/m³ with essentially monodisperse drop diameters of 100 and 200 microns. Moreover, the generators injected the drops into the FITS chamber with a gentle sidewise direction.

Next, we began a series of shakedown tests to determine the effect of these suspended water drop densities on hydrogen burns of 6.5 and 10 percent. During these shakedown tests, it became apparent that our ability to achieve predetermined concentrations of hydrogen in air was poor. Although some of the uncertainty arose from our analytical procedures, a significant fraction arose from our dosing procedures. Here we used the pressure-volume product; where the volume was large--5.6 m³, the volume of the FITS chamber--and pressure increments were small. The pressure measurements were determined with a Bourdon gauge attached to the chamber with approximately 50 m of 6-mm copper tubing that led to our control room.

In order to reduce these uncertainties, we improved both our analytical procedures (mass spectrometry) and our gas dosing technique. To achieve the latter, we installed a system for dosing in which a new approach to the premeasurement was made. In this system, we used a small volume, a 9-liter gas cylinder, and a large pressure, measured with a Bourdon gauge that operated up to 500 psig maximum. This gauge was located less than a meter away from the gas cylinder. With this new system, we were able to achieve the following results: In 6 experiments with a target composition of

10 percent hydrogen in air, we achieved an analyzed composition of 9.64 ± 0.07 percent. In 13 experiments with a target composition of 6.5 percent hydrogen in air, we achieved an analyzed composition of 6.29 ± 0.13 percent. Note that these analyzed compositions combine both FITS chamber filling and analytical errors. We suspect that the apparent systematic error shown in these results may be due to an incorrect estimate of the volume of the FITS chamber, which is normally cited to be 5.6 m^3 ; perhaps due to incomplete accounting for the many ports and side arms, the true volume may be somewhat higher.

Our experiments with the combustion of lean hydrogen-air mixtures in the presence of monodispersed water drops in the FITS chamber have been concluded. We studied two target concentrations of hydrogen in air, 10 and 6.5 percent, which were above and below the downward propagation limit of 9 percent, respectively.

Control experiments were performed in a quiescent mode without sprays in both mixtures. At 10 percent, the pressure-time traces were repeatable from shot to shot, as shown in Figure 11. Moreover, essentially all hydrogen was consumed during the burns. However, at 6.5 percent, the pressure traces were not reproducible shot to shot (traces at the right in Figure 12). Also, the combustion of hydrogen was incomplete, with only about 28 percent burned. These observations generally agree with the following earlier observations:²

1. Above the downward propagation limit, hydrogen burns globally (i.e., in all directions), consuming essentially all hydrogen in the chamber and producing reproducible peak pressures that approach the adiabatic isochoric complete combustion (AICC) limit.
2. Below this limit, hydrogen burns incompletely with an upward, approximately conical, flame which yields variable pressures traces with peak pressures that fall well below the AICC limit (Figure 13 which is taken from Figure 26 in Reference 2).

Although our data for the burns with water drops are not fully analyzed, we have made these preliminary observations:

- 1'. For 10 percent mixtures of hydrogen in air burned in the presence of 100 micron-diameter water drops at volumetric densities of approximately 100 g/m^3 , the peak pressures and fraction of hydrogen consumed during the burn were unchanged compared to control burns, while the rates of both pressure rise and decay increased as shown in Figure 11; the normalized peak pressure $\Delta P/P_0$ was approximately 3.7.

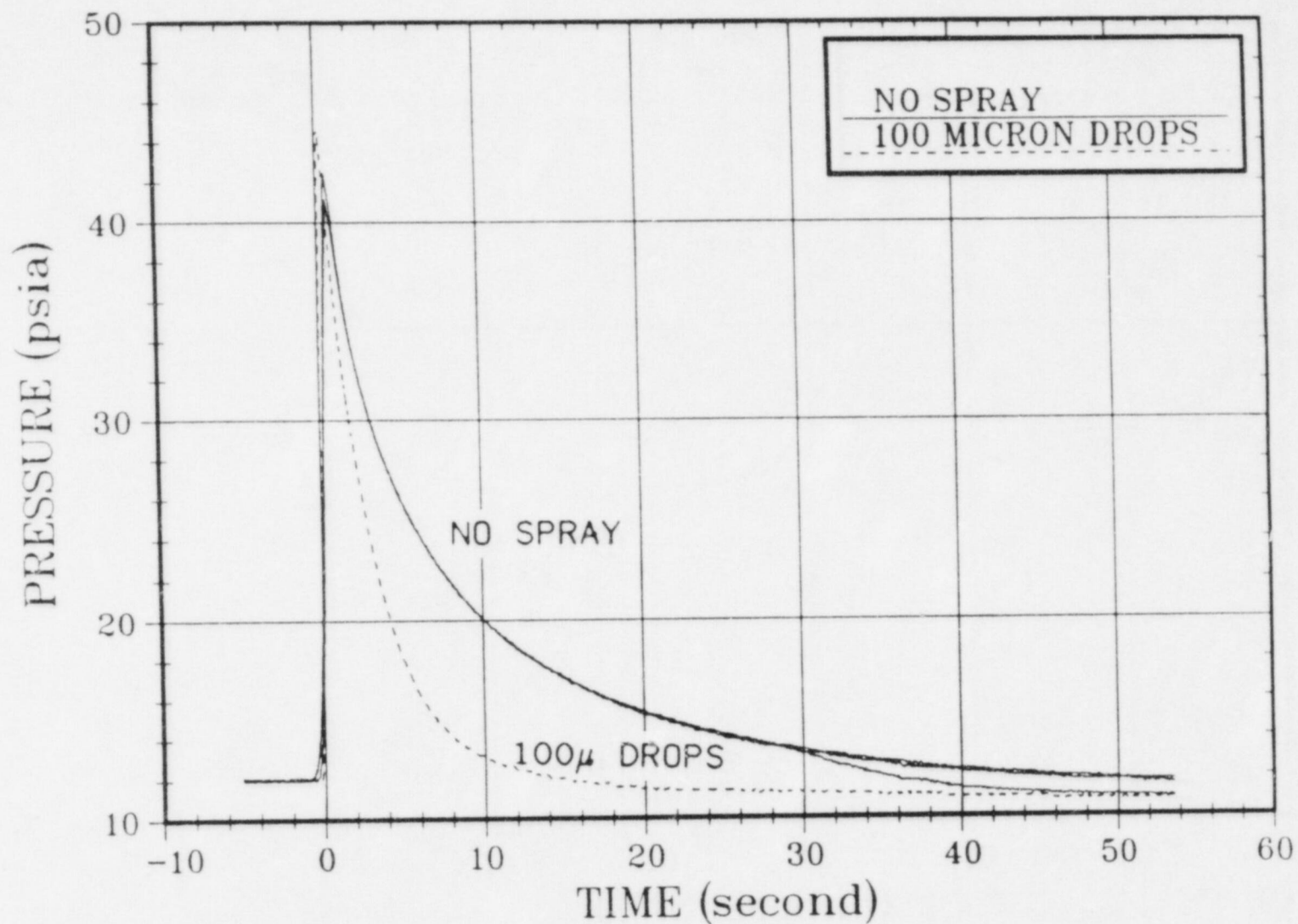


Figure 11. Pressure-Time Traces Recorded During the Combustion of 10 Percent Hydrogen-in-Air Mixtures in the 5.6-m³ FITS Chamber. The solid traces were recorded when the gas mixtures were dry; the dashed traces were recorded when 100 micron-diameter monodisperse water drops at a volumetric density of ~100 g/m³ were dispersed in the gas mixtures.

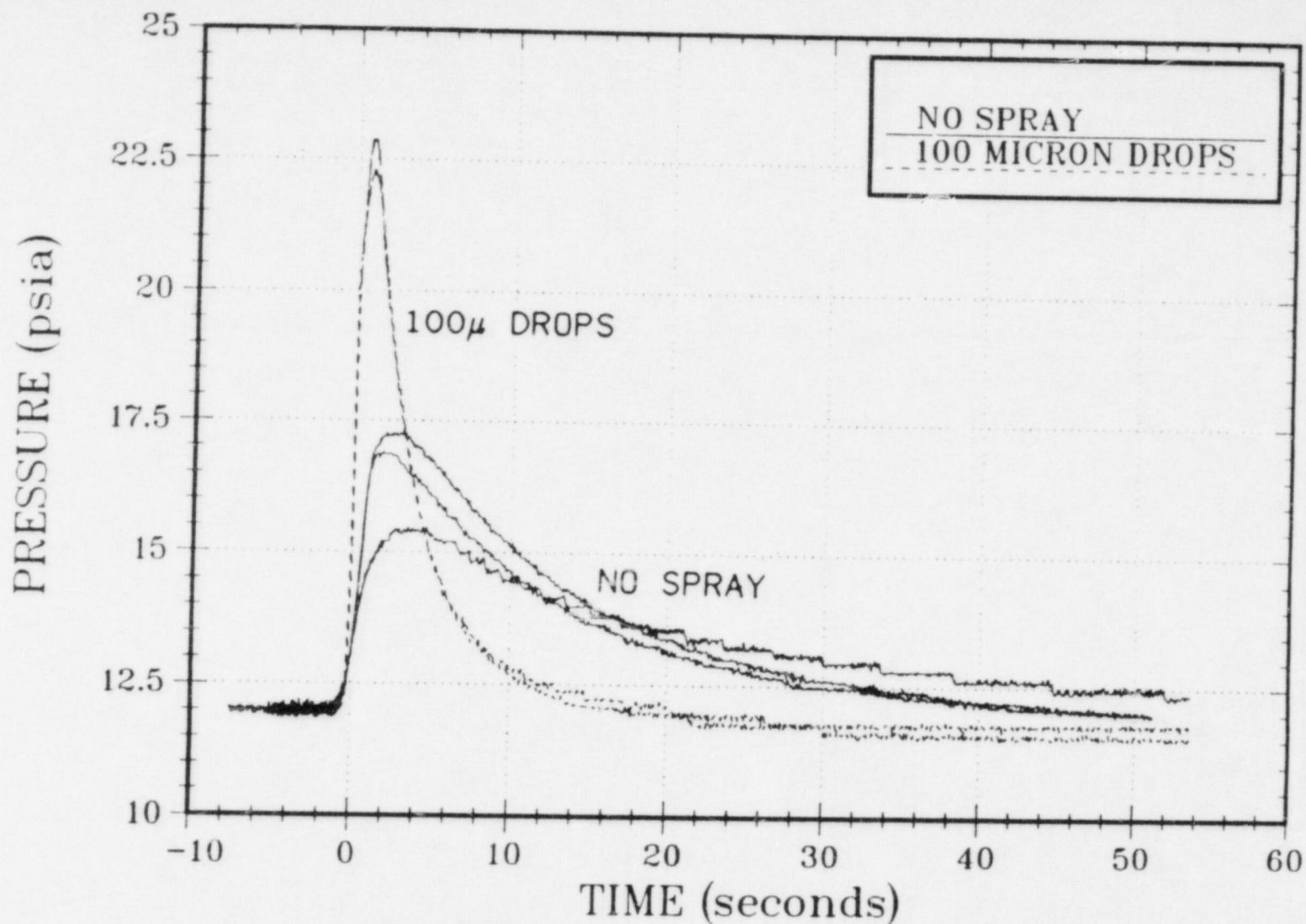


Figure 12. Pressure-Time Traces Recorded During the Combustion of 6.5 Percent Hydrogen-in-Air Mixtures in the 5.6-m³ FITS Chamber. The solid traces were recorded when the gas mixtures were dry; the dashed traces were recorded when 100 micron-diameter monodisperse water drops at a volumetric density of ~100 g/m³ were dispersed in the gas mixtures.

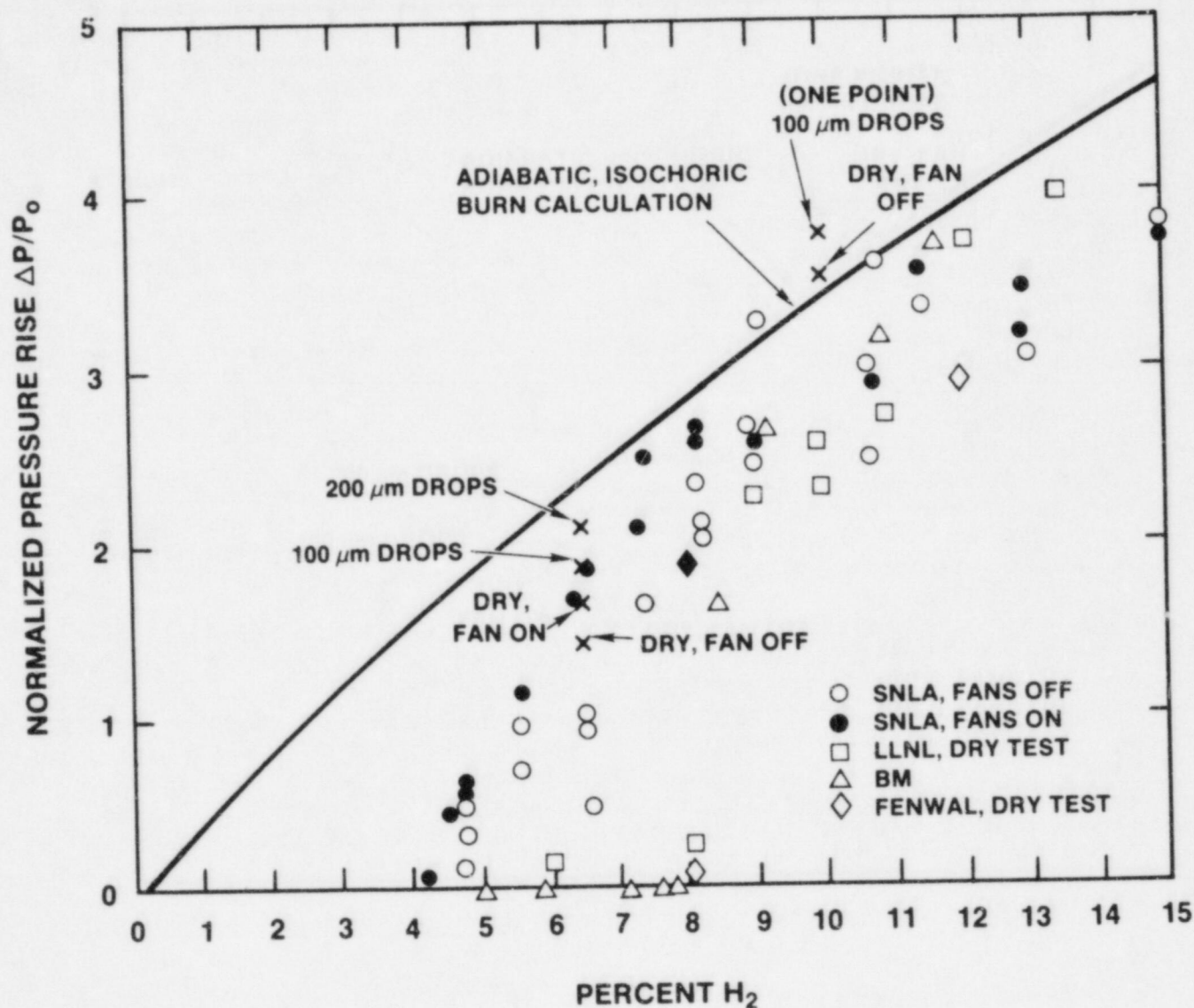


Figure 13. Normalized Pressure Rises Produced With and Without Monodisperse Water Drops and Fan Operation in 6.5 and 10 Percent Hydrogen-Air Burns in the VGES Chamber Compared With Earlier Experiments. For original plot and explanations of laboratory code, see Figure 26 of Reference 2.

- 2'. For 6.5 percent mixtures of hydrogen and air burned in the presence of 100 micron-diameter water drops at volumetric densities of 100 g/m^3 , the fractions of hydrogen burned approximately doubled, rising from 28 to 55 percent, while the pressure traces became reproducible with peak pressure rises also approximately doubled above the control experiments as shown in Figure 12; here, the normalized peak pressure $\Delta P/P_0$ was approximately 1.9 and rates of both pressure rise and fall were increased substantially.

The action of water drops described in (2') and shown in Figure 13, differs somewhat from the view that water sprays create "turbulence" or "stirring" during lean hydrogen burns. Although these quantities are seldom quantified, they are commonly used to explain the generally more vigorous burns observed with sprays at hydrogen concentrations below the downward propagation limit of 9 percent (Reference 12 describes the action of sprays in the NTS large-volume burns).

Our experiments suggest that the effect of sprays may have more to do with the presence of heterogeneous water (the drops) dispersed throughout the combustible gases than to the gas motion produced by the falling drops. This statement is based on the observation that the vigor of the lean burns increases comparably in both the NTS experiments and in our FITS experiments even though the gas motion in FITS is perhaps an order of magnitude lower than in the NTS experiments. We can estimate roughly from the calculations by Marx⁹ that gas motion induced by the coarse sprays generated by standard containment nozzles (diameters from ~ 20 to $2000 \mu\text{m}$) in the NTS chamber might have velocities of 5 or 10 m/s, with much recirculation and shear flow, while in FITS, the gentle, sidewise injection of our $100 \mu\text{m}$ -diameter drops might induce gas velocities only on the order of the terminal velocity of the drops: namely, about 0.25 m/s.¹³

1.5 Nonpowered Igniters for Lean Hydrogen-Air Mixtures (L. R. Thorne, J. V. Volponi, and W. J. McLean, 8353)

A loss-of-coolant accident (LOCA) at a light water nuclear reactor may rapidly produce large quantities of hydrogen gas. The hazards associated with the resulting hydrogen-air mixture may be reduced by intentionally burning it at sufficiently low hydrogen concentrations that little, if any, damage to the containment will occur. Current implementations of this strategy make use of glow plugs that require an uninterrupted supply of electrical power. In the event of a serious accident, however, electrical power may not be available. Thus there is a need for a nonpowered igniter.

For any igniter to be a practical safety device, it must also operate reliably under the conditions which might exist during an LOCA. These include high temperature, high humidity, wind velocities from 0 to 10 m/s, and the presence of water spray and steam.

We have developed an igniter^{1*} that can operate under most of these conditions and that does not require an external source of power of any kind. The igniter is composed of a catalytic substrate and several wires which project into the unreacted gas. The substrate is an alumina honeycomb (4.4-cm diameter and 3-cm height, with 0.2-cm diameter cells) that is coated with high-surface-area platinum particles to about 1.7 weight percent platinum. The wires are also platinum (0.0123-cm diameter and 4-cm long).

The igniter operates by catalyzing the reaction between hydrogen and oxygen on the platinum surface. This reaction produces heat that increases the platinum surface temperature and causes the reaction rate to accelerate. Further reaction increases the temperature of the device to the point that gas phase ignition occurs. Because of its high platinum surface area, the honeycomb starts to heat first when the device is initially exposed to a lean hydrogen-air mixture. By itself, the honeycomb will not get hot enough to cause ignition at low hydrogen concentrations because the reaction rate is limited by the transport rate of hydrogen to the surface. The honeycomb, however, provides a "thermal boost" to the wires which otherwise would not heat because of their low effective surface area. Once boosted, the wires heat to a higher temperature than the honeycomb because their shape provides for better transport of the reactants to their surface. Thus, gas phase ignition is initiated on the surface of the wires.

Our laboratory tests show that ignition occurs after a typical induction time of 20 to 400 s. This induction time depends on the hydrogen concentration, gas flow velocity, gas temperature, and the relative humidity. We have measured induction times for hydrogen concentrations in the range of 5.5 to 11 percent, gas flow velocities between 1.7 and 19.5 cm/s, gas temperatures between 20° and 65°C, and relative humidities between 5 and 98 percent. We have found that induction times are shorter for mixtures with higher hydrogen concentrations, higher flow velocities, higher gas temperatures, and lower relative humidity.

* The design of the optimized catalytic igniter described herein is currently the subject of a Sandia patent disclosure.

In addition, the igniter operates repeatedly. Some of the igniters used in this study were cycled tens of times without any sign of reduced performance. This is a desirable characteristic since the hydrogen produced during a LOCA may require several separate ignition events to eliminate hazardous concentrations of hydrogen. Liquid water blocks the catalytic sites and defeats the igniter. But, when a wet igniter is dried it operates normally, indicating that water does not poison the catalytic sites. Preliminary tests indicate that liquid water may not defeat igniters made with "wet proofed" catalytic substrates developed at Atomic Energy of Canada's Chalk River Laboratories. However, higher hydrogen concentrations are required for ignition to occur.

Most recently, we have tested the catalytic igniters at the FITS facility (an instrumented, 1.5-m diameter by 4-m tall high-pressure tank located at Sandia). These tests were performed in collaboration with the Severe Accident Response Division (6427). This was the first time the igniter was tested in a static atmosphere. These tests are important because previous laboratory tests showed that the ignition induction time increases significantly for flow velocities below 10 cm/s and static conditions may in fact prevail in a LOCA in which the igniter must perform properly.

The FITS tests proceeded by filling the tank with a predetermined amount of hydrogen to give the desired hydrogen concentration. Then the tank was stirred with a pneumatic fan for 10 min, then allowed to stand for 10 min. The igniter was then inserted from a side arm of the tank through a 10-cm diameter ball valve into the center of the tank. The side arm was filled with helium to prevent preignition when the igniter was inserted. Eight tests were performed using either 10 or 6.5 percent hydrogen. The typical induction time was less than 90 s, even for a 10 percent hydrogen mixture in which the relative humidity was nearly 100 percent. Four of the tests had longer induction times but in several of these tests the igniter may have been rotated so that the honeycomb cells were not vertically oriented, thereby reducing the flow through the catalytic substrate which is aided by buoyancy if the cells are vertical.

The platinum igniter seems to produce burns identical to glow plug igniters in the FITS chamber. This is illustrated by the two pressure traces shown in Figure 14 for 10 percent hydrogen-in-air mixtures.

These tests demonstrate that a nonpowered igniter for lean hydrogen-air mixtures is in fact feasible and that it could contribute to the safety of light water nuclear reactors by its ability to operate under the conditions present during a LOCA, including loss of electrical power.

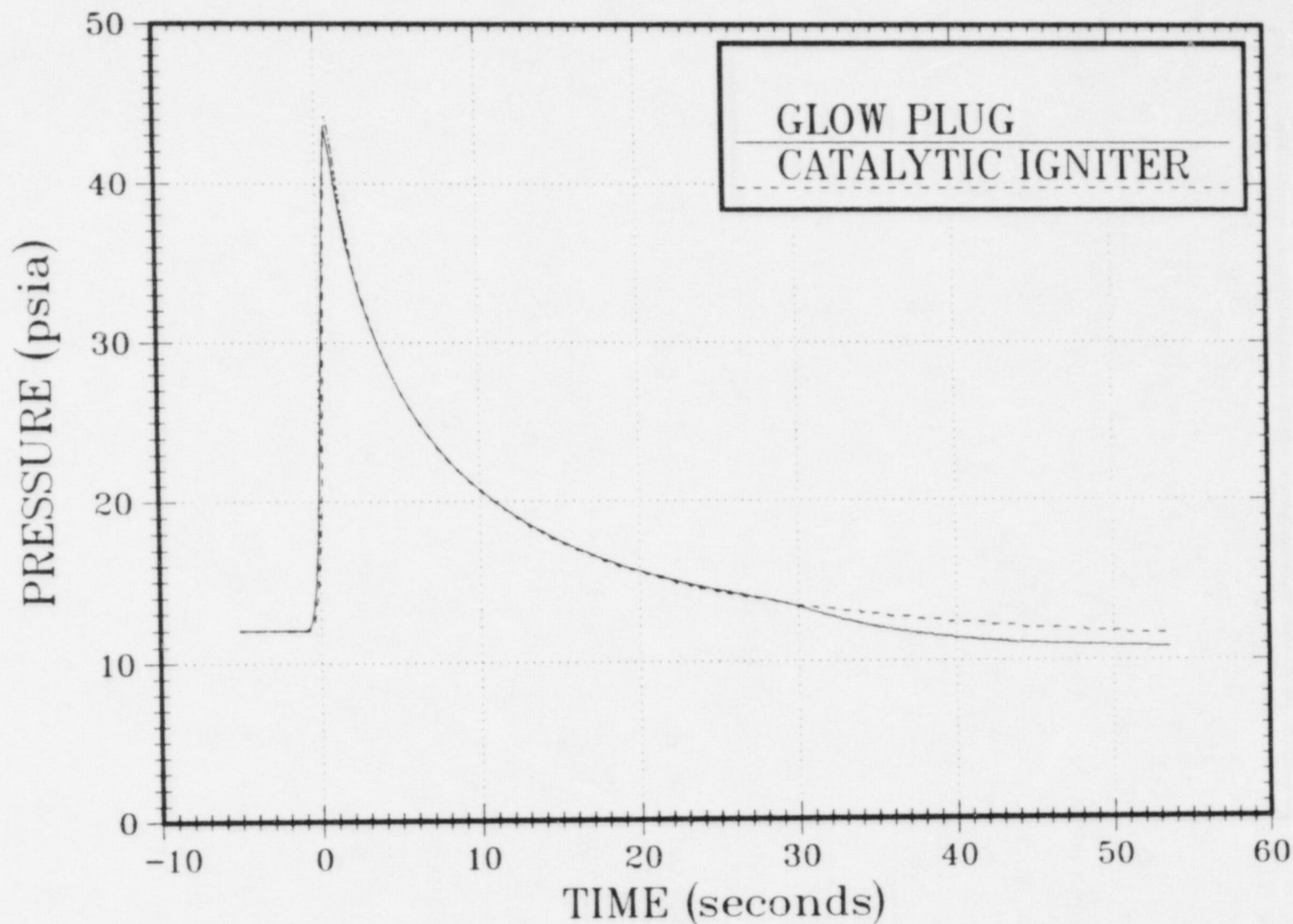


Figure 14. Comparison of Pressure-Time Traces Recorded During Burns of 10 Percent Hydrogen-in-Air Mixtures in the 5.6 m³ FITS Chamber. Burns were initiated with a glow plug igniter (solid trace) and a platinum catalytic igniter (dashed trace).

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