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June 1970

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DEVELOPMENT OF A CONTINUOUS SAMPLER-MONITOR
FOR THE LOFT CONTAINMENT ATMOSPHERE

R. E. Schindler, R. R. Hammer
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Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

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IN-1309
Issued: June 1970
Instruments
TID-4500

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ACKNOWLEDGMENTS

The development of the Continuous Sampler-Monitor represents a cooperative effort involving numerous individuals. Dr. C. M. Slansky is due credit for the original concept of the Continuous Sampler-Monitor and also provided initial program guidance. Dr. D. H. Walker and personnel of the LOFT and Radiological Experiments Section formulated the CSM performance criteria and provided program guidance. Mr. L. O. Johnson of the Instrument Development Branch designed the liquid control system for the separator. Messrs. J. H. Keller and W. J. Maeck of the Chemical Research Section developed the sorbent components for the P-I Samplers and the chromatographic techniques used to evaluate the CSM. The Analytical Chemistry Branch performed the many analyses required in the development program. Special appreciation is also due Messrs. W. P. Palica, R. L. Nebeker, and R. L. Ring (Reactor Chemical Applications Section) who operated the Contamination-Decontamination Experiment in which most of the development testing was performed.

ABSTRACT

A Continuous Sampler-Monitor (CSM) has been developed to determine fission-product concentrations in the containment building atmosphere during the LOFT Integral Test Program. The specific objectives of the CSM installation in LOFT are to remotely sample and determine during the test, the airborne noble gas, total iodine, and organic iodide concentrations in the containment atmosphere, and to monitor changes in total activity in the containment atmosphere.

A small stream (1 to 3 liters/min) of the atmosphere is continuously drawn into a scrubber where the elemental iodine and most of the particulate fission products are washed into a stream of basic-thiosulfate scrubbing solution leaving noble gases, organic iodides, and hypiodous acid in the gas stream. The scrubbed gas stream and the scrubbing solution flow in a common line to a sampling station located outside the containment building where they are separated. The gross activity levels of the separated streams are monitored, and samples of each stream are analyzed to determine the concentrations of individual nuclides in the containment atmosphere.

In tests conducted with low concentrations of iodine and xenon in an 86-ft³ vessel under simulated LOFT atmospheric conditions, the CSM accomplished its objectives. Xenon values from the CSM were representative within 4% of xenon concentrations in the containment atmosphere. The CSM total iodine values were accurate to within 50%, and the CSM organic iodide values were accurate to within 30% when corrected for the methyl iodide absorbed in the scrubbing solution. The gross activity monitors provided reasonable gross activity records from which the time of an activity release could be determined with an accuracy of 2 or 3 minutes.

Further testing is recommended to assure the CSM will perform satisfactorily on the short-cooled, mixed fission products expected in LOFT.

SUMMARY

A Continuous Sampler-Monitor (CSM) has been developed to determine fission product concentrations in the containment building atmosphere during the LOFT Integral Test Program. The specific objectives of the CSM installation in LOFT are to remotely sample and determine during the test the airborne noble gas, total iodine, and organic iodide concentrations in the containment atmosphere, and to monitor the total activity in the containment atmosphere.

A small stream (1 to 3 liters/min) of the atmosphere is continuously drawn into a scrubber where the elemental iodine and most of the particulate fission products are washed into a stream of basic-thiosulfate scrubbing solution, leaving noble gases, organic iodides, and hypiodous acid in the gas stream. The scrubbed gas stream and the scrubbing solution flow in a common line to a sampling station located outside the containment building where they are separated. The gross activity levels of the separated streams are monitored, and samples of each stream are analyzed to determine the concentrations of individual nuclides in the containment atmosphere.

The CSM was evaluated in the 86 ft³ CDE vessel under simulated LOFT atmospheric conditions with low concentrations of xenon, methyl iodide, and an iodine mixture containing mostly elemental iodine. Additional data were obtained from laboratory tests with methyl iodide and fluorescein dust.

The noble gases--xenon and krypton--present few sampling problems. Except for a small fraction--expected to be less than 1%--which dissolves in the scrubbing solution, the noble gases remain in the CSM scrubbed gas stream where their concentrations can be determined by analysis of gas stream samples. In CDE tests with xenon-133, the xenon concentrations determined from CSM samples agreed within 4% with xenon concentrations of samples taken directly from the containment atmosphere.

In CDE "elemental" iodine runs with initial iodine concentrations from 0.05 to 7 $\mu\text{g}/\text{m}^3$, the CSM provided total iodine concentrations accurate to within $\pm 50\%$ when compared with concentration values obtained

with the Particulate and Iodine (P-I) Sampler. In two runs, the total iodine concentrations determined with the CSM and the P-I Samplers agreed within $\pm 10\%$.

The concentrations of organic iodides in the containment atmosphere were determined by correcting the CSM gas stream organic iodide concentration, analyzed by chromatographic separation of the organic iodides followed by gamma-ray spectrometry, for the fraction of the organic iodide absorbed by the scrubbing solution. When the CSM was operated at the recommended conditions--a gas sampling rate of 2 liters/min and a scrubbing solution containing $1M$ sodium hydroxide and $0.007M$ sodium thiosulfate--from 10 to 50%, depending on temperature, of the methyl iodide was absorbed by the scrubbing solution. After correcting for the average absorption, the CSM provided organic iodide concentrations accurate to $\pm 30\%$.

The gross activity monitors on the CSM gas and liquid streams provided gross activity traces in CDE elemental and methyl iodide runs which agreed with the expected behavior. There was a time lag of ~ 5 min between the time of sampling and the time the activity was detected by the monitors due to the time required to transport the sample through the long lines needed for the LOFT installation. The activity traces obtained in elemental iodine and methyl iodide tests indicate that by correcting for a time lag of about 5 min, the time of a sudden activity release can be determined with an accuracy of 2 or 3 min with the CSM gross activity monitors.

Based on the criteria developed for the LOFT Integral Test Program, the CSM accomplished its objectives in the low concentration releases of a single nuclide (either iodine-131 or xenon-133). However, prior to LOFT, reliable CSM performance should be demonstrated for a fission product mixture, including short-lived nuclides and particulates, released from an irradiated fuel pin under LOFT-ECC conditions. Tests in a large containment vessel are also needed to characterize the CSM performance, because the rapid washout of elemental iodine vapors in the 86 ft^3 CDE vessel limited the evaluation of the CSM performance on elemental iodine.

Recommendations are also made for design evaluations and modifications of CSM components on which improvements are desirable.

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DEVELOPMENT OF A CONTINUOUS SAMPLER-MONITOR FOR THE LOFT CONTAINMENT ATMOSPHERE

I. INTRODUCTION

The LOFT Integral Test Program consists of a series of engineering-scale tests, each simulating a reactor loss-of-coolant (LOC) accident, in which Emergency Core-Cooling Systems are tested^[1,2]. In LOFT, the coolant will be released from a 55 MW pressurized water reactor, permitting the core to overheat and release fission products into the reactor vessel and 300,000-ft³ containment building. The increase in the fuel temperature will be terminated by activation of an emergency core-cooling system prior to melting of the fuel pins. However, even with a core-cooling system, the fuel temperatures reached will probably result in some cladding failures and subsequent release of small quantities of fission products.

The atmosphere in the LOFT containment building following the coolant blowdown will consist of an air and steam atmosphere containing a short-cooled fission product mixture released from failed fuel pins. The character of the fission-product mixture will depend upon the extent of fuel pin damage. If the damage is limited to cladding failure, the fission-product mixture will consist mainly of the noble gases and their daughters, and lesser quantities of the radioiodines as elemental iodine vapors, organic iodides, iodine adsorbed on particulates, and hypiodous acid vapors. Particulate fission products, such as cesium, tellurium, and molybdenum, would be present if the fuel pins overheat sufficiently to melt or vaporize. The time and space dependent fission-product concentrations and compositions in the LOFT containment atmosphere will be determined by two samplers--the Sequential Particulate and Iodine (SP-I) Sampler and the CSM. The SP-I Sampler is an assembly of cartridges (often referred to as "Maypacks"), each containing a series of filters to collect particulate fission products, and adsorber beds to collect the various volatile iodine species. SP-I Samplers located inside the containment building will be sampled remotely during the test and removed from the containment building after the test. Analysis of the

sorbent beds and filters will then provide data on the atmospheric total iodine concentration, the concentrations of the various iodine species, and the concentrations of particulate fission products. The CSM uses a scrubber located in the containment building to continuously and remotely sample the atmosphere and separate the fission products into liquid and gas streams, which leave the containment building as shown in Figure 1. Samples provided by the CSM during the LOFT will be analyzed to determine the noble gases, total iodine, and organic iodide concentrations in the containment atmosphere. The total iodine and organic iodide concentrations will be measured by analysis of samples from both the SP-I and CSM Samplers; however, the CSM will provide samples while the test is underway. The concentrations of particulate fission products and the differentiation between the chemical forms of iodine will be provided by the SP-I Samplers only. The noble gas samples will be provided by the CSM only. The CSM will also continuously monitor and record the gross activity levels in the containment atmosphere.

The CSM was tested in the pilot-plant-scale Contamination-Decontamination Experiment (CDE)^[3] under simulated LOFT atmospheric conditions with tracer releases of xenon, methyl iodide, and a mixture of elemental iodine vapors with small quantities of organic iodides and hypoiodous acid. This report summarizes the development status of the CSM at the end of 1969. The development effort by Idaho Nuclear Corporation on the Continuous Sampler-Monitor has been terminated, and the CDE facility has been shut down. Plans to continue testing of the CSM in the Containment Research Installation (CRI) at ORNL are being considered.

Fig. 1 Schematic diagram of Continuous Sampler-Monitor

II. DESCRIPTION OF CSM AND TEST FACILITIES

The performance of the CSM was evaluated using both the pilot-scale CDE facility and a laboratory mockup. In the CDE, performance was evaluated by comparing airborne activity concentrations from the CSM with those determined by other atmosphere samplers. The CSM total iodine concentrations were compared with those determined by the P-I Samplers; the CSM noble gas and organic iodide concentrations were compared with those determined by the Raw (or Filtered) Gas Samplers. During the development program, the CSM was tested in the CDE with mixed fission products released from irradiated fuel pins, and tracer-level noble gas, elemental iodine, and methyl iodide releases. The laboratory tests were conducted to determine the CSM performance for methyl iodide and fluorescein dust. The following sections describe the CSM, the laboratory and CDE test facilities, and the samplers used to evaluate performance of the CSM.

1. CSM DESIGN, OPERATION, AND SAMPLE ANALYSIS

1.1 CSM Design

Elemental iodine vapor cannot be transported in a gas stream through lines of any significant length, because it deposits on the wall of the sampling line. The CSM is designed to minimize iodine deposition by removing the elemental iodine from the gas stream with a scrubber located inside the containment building at the point of sampling as shown in the simplified flowsheet portrayed in Figure 2. A detailed CSM description and flowsheet are given in Appendix A. A continuous sample of the containment atmosphere is drawn into a 2-in. OD by 12-in. long "Teflon" (TFE) scrubber (shown in Figures 3 and 4) by a pump located outside the containment building and connected to the scrubber by a long length of 3/8-in. diameter stainless steel line. The atmosphere sample enters the vertical scrubber through ports at the top of the scrubber and is washed by a fine mist of scrubbing solution sprayed at a controlled rate through a pressure-atomizing spray nozzle. Iodine and particulate-associated fission products are absorbed into the scrubbing solution,

Fig. 2 Simplified flowsheet of Continuous Sampler-Monitor.

Fig. 3 Cutaway view of CSM scrubber.

Fig. 4 CSM installed in CDE containment vessel.

and the iodine is converted to a soluble form. The gas sample stream, carrying the liquid with it, flows from the outlet at the bottom of the scrubber through a long length (100 to 200 ft) of 3/8-in. OD stainless steel tubing to a cooler located outside the containment building where excess moisture is condensed from the gas. After being cooled, the gas and the scrubbing solution flow to a pot where the liquid is separated from the gas. The scrubbing solution and gas then flow separately to gross activity monitors and sampling stations.

A liquid-level control system--consisting of a differential-pressure transmitter, a controller, and a variable-speed metering pump--measures the liquid level and pumps the solution out at a rate which maintains a constant liquid level in the separator pot. The atmospheric sample flow is controlled downstream of the gas sampling station by a control system which maintains a constant mass flow of dry air. This gas flow control system also maintains a constant volumetric flow rate of the steam-air mixture into the scrubber, as long as the air and steam in the containment building are well mixed. A vacuum pump downstream of the gas flow control system maintains the pressure difference which drives the gas flow.

In an earlier CSM design, which was tested in the three CDE runs with irradiated fuel pins, the gas and liquid streams separated inside the scrubber and discharged from the scrubber through separate lines which carried the scrubbed gas and scrubbing solution outside the containment vessel. Problems with moisture condensation in the gas line were encountered with this earlier design. The current CSM design was adopted because the gas and liquid streams are easier to separate outside the containment vessel than inside the scrubber, and because cooling the gas and liquid streams before separation reduces moisture carry-over into the gas stream.

1.2 Sampling and Analysis of CSM Effluent Streams

Evaluation of the CSM performance was based primarily on the analyses of samples from the gas and liquid effluent streams. The concentrations of individual nuclides in curies per unit volume of containment atmosphere were calculated from the sample analysis and the gas and

liquid stream flow rates. The total iodine activity in the CSM liquid stream was determined by analysis of liquid samples collected intermittently in small bottles and analyzed by gamma-ray spectrometry. The total iodine concentration in the CSM gas stream was measured by analysis of small charcoal packs (Barnebey-Cheney Type 727 charcoal) through which a known volume of gas stream was diverted. The iodine collected on the charcoal bed was then determined by gamma-ray spectrometry. The total iodine concentration in the containment vessel was calculated by multiplying the iodine concentration in the liquid stream by the liquid-to-gas-flow ratio and adding this value to the total iodine concentration in the gas stream.

The concentrations of light organic iodides (methyl, ethyl, and propyl iodides) and noble gases in the CSM gas stream were determined by analysis of gas samples collected intermittently using gas sample cylinders which were flow purged with the gas being sampled. The light organic iodide or xenon fraction in the gas sample was separated and purified by a chromatographic technique*, then collected in charcoal beds, and finally analyzed by gamma-ray spectrometry. During the xenon tracer runs, the gas sampling cylinders were also counted directly by gamma-ray spectrometry to obtain xenon concentrations for comparison with those obtained using the chromatographic separation technique.

1.3 CSM Operating Conditions

CSM operating conditions were varied during the testing program to determine their effect on the CSM performance. The scrubbing solution flow rates used in the tests ranged from 60 to 200 ml/min, while the gas sampling rates ranged from 200 to 3100 std ml/min. The latest tests, which provided the best results, were conducted at a solution flow of 110 ml/min and with two different gas sampling rates. The low gas sampling rate ranged from 800 to 1000 ml/min, while the high gas sampling rate ranged from 2000 to 3100 ml/min. With the exception

*The organic iodides and xenon were separated from the inorganic iodine species by a Beckman GC-5 chromatograph with a 0.25-in. diam column of Porapak Q preceded by a silica gel plug to retain inorganic iodides. The xenon samples were separated with 14-ft long column operated 25°C with a 50 ml/min flow of helium carrier gas. The organic iodide samples were separated in a 4-ft long column operated at 150°C with a 100 ml/min flow of helium carrier gas.

of Tracer Run 4, scrubbing solutions were sodium hydroxide solutions or sodium hydroxide and sodium thiosulfate mixtures. The operating conditions for each run are tabulated in Table I.

2. LABORATORY TEST FACILITIES

Laboratory test facilities were used when feasible to supplement the CDE tests. The laboratory test systems which provided organic iodide and particulate scrubbing data are described in the following sections.

2.1 Laboratory System for Evaluating Organic Iodide Scrubbing

A laboratory model of the CSM, without the scrubber, was installed to evaluate the effect of scrubbing solution composition on the absorption of methyl iodide in the CSM scrubbing solution. The laboratory test system consisted of a 100-ft coil of stainless steel tubing through which scrubbing solution and gas flowed cocurrently. The spray scrubber was omitted because most of the CSM gas-liquid contact area (which controls the methyl iodide adsorption) is in the 100- to 200-ft LOFT discharge line through which the atmosphere sample and scrubbing solution flow cocurrently.

A flowsheet of the laboratory apparatus is shown in Figure 5. Methyl iodide or ethyl iodide was generated by reacting a basic solution of sodium iodide (the iodide was 5 to 20 atom percent iodine-131 with the balance iodine-129 and stable iodine-127) with dimethyl or diethyl sulfate. The organic iodide was then mixed with the air sample stream ahead of the coil. To determine the inlet methyl iodide concentrations, a small fraction of the gas stream was bled through a charcoal bed sampler at a measured rate, and the sampler was analyzed for iodine. The gas stream was then mixed with scrubbing solution, which had been preheated to the test temperature, and passed through the 100-ft coil where the organic iodides were absorbed by the scrubbing solution. The scrubbing solution discharging from the coil was separated (by settling) from the gas stream and drained into a holding tank which was sampled. The scrubbed gas stream flowed through a charcoal pack sampler which

TABLE I
CSM OPERATING CONDITIONS

Test	Type of Activity	Gas Sample Flow (ml/min)	Scrubbing Solution Flow (ml/min)	Scrubbing Solution Composition	
				NaOH (M)	Na ₂ S ₂ O ₃ (M)
Fuel Pin Run 3	Mixed Fission Products	260	60	0.5	0.5
Fuel Pin Run 4	Mixed Fission Products	200	200	0.5	0.5
Fuel Pin Run 5	Mixed Fission Products	180	200	0.5	0
Tracer Run 1	Xenon (Xe-133)	210	110	0.1	0
Tracer Run 2	Xenon (Xe-133)	210	110	0.1	0
Tracer Run 3	Iodine (I-131)	365	110	0.1	0
Tracer Run 4	Iodine (I-131)	430 to 550	110	0.08	0.00005 ^[a]
Tracer Run 5	Methyl Iodide (I-131)	1000	110	1.14	0.012
Tracer Run 6	Iodine (I-131)	800 or 2000	110	0.92	0.14
Tracer Run 7	Methyl Iodide (I-131)	1000 or 2000	110	0.092	0.010
Tracer Run 8	Methyl Iodide (I-131)	1000 or 2000	110	1.0	0.005
		1000 or 2000	110	0.75	0.07
Tracer Run 9	Iodine (I-131)	1000 or 2000	110	1.0	0.007
Tracer Run 10	Iodine (I-131)	800 or 2000	110	1.0	0.007
Tracer Run 11	Iodine (I-131)	800 or 3100	110	1.0	0.01

[a] Na₂SO₃ rather than Na₂S₂O₃.

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Fig. 5 Schematic flowsheet of laboratory mockup of CSM for testing on organic iodides.

collected the incident iodine in the gas stream and through a gas volume meter. The fraction of organic iodide absorbed was calculated from the relative amounts of iodine in the gas and liquid streams leaving the scrubbing coil. The fractions of methyl and ethyl iodide removed from the gas by sodium hydroxide-sodium thiosulfate mixtures of varying concentrations were determined in the test system at 80°C and a gas flow of 1000 ml/min.

2.2 Laboratory System for Evaluating Particulate Scrubbing

Laboratory tests were also conducted to evaluate the scrubbing efficiency of the initial CSM concept. In this system, the scrubbed gas and scrubbing solution streams separated in the scrubber instead of flowing cocurrently through the common discharge line used in the current design. A schematic flowsheet of the test apparatus is shown in Figure 6. The fluorescein dust was produced by atomizing a fluorescein solution and drying the resulting spray. Air containing the fluorescein dust was passed through the scrubber at rates from 160 to 650 ml/min and scrubbed by a spray of water from a small pressure-atomizing spray nozzle* operating at the normal CSM spray pressure of 500 psig. The scrubber performance was measured by collecting, with glass filters, the fluorescein content of measured samples of the air entering and leaving the scrubber. A colorimetric method of analysis was used to measure the quantity of fluorescein on each filter.

The fluorescein particles ranged in size from 0.05 to 0.7 micron diameter and had a mass median diameter of 0.43 micron. The particle size distribution shown in Figure 7 was obtained from electron microscope photographs of two samples of the fluorescein dust.

3. CDE TEST FACILITIES

3.1 Contamination-Decontamination Experiment

The Contamination-Decontamination Experiment (CDE) is a pilot-plant-scale facility which produces high-level fission product aerosols

*Four nozzle sizes were tested: Delavan nozzle sizes 0.50 gph-90°, 0.75 gph-70°, and 1.00 gph-70°, and Spraying Systems nozzle size 0.60-1/4 LNN.

Fig. 6 Schematic flowsheet of apparatus for fluorescein scrubbing test.

Fig. 7 Particle size distribution of fluorescein dust.

or tracer-level aerosols under conditions simulating those expected in LOFT. A cutaway drawing of the CDE is shown in Figure 8. In the CDE tests, the containment atmosphere was sampled continuously with the CSM and periodically with the other atmosphere samplers--the P-I and Raw Gas Samplers--whose total iodine and organic iodide concentrations were compared with the CSM values.

Two methods--fuel pin meltdowns and tracer-level injections--were used to produce fission product aerosols in the containment vessel. One fission product aerosol was generated by melting an irradiated fuel pin in the presence of steam, using an induction furnace. The fission products released were transported by steam to an 86-ft³ containment vessel in which conditions were controlled to simulate anticipated LOFT conditions. Additional details are reported elsewhere^[3]. The tracer level aerosols were produced by injecting xenon, elemental iodine or methyl iodide into the containment atmosphere rather than fission products from a fuel pin. Iodine-131 as elemental iodine was prepared by drying a basic solution of sodium iodide containing 5 to 20% iodine-131 with the balance as iodine-129 and stable iodine-127, then oxidizing the residue with molten potassium dichromate. Methyl iodide traced with iodine-131 was prepared by reacting dimethyl sulfate with iodine-131 traced sodium iodide. Xenon-133 (a gas) was injected directly from the ampule in which it was received. A glass ampule containing the desired nuclide was broken in an injection tube, and the nuclide was swept into the CDE vessel by argon. Neither carrier iodine nor carrier xenon was added in any of the tests.

3.2 Particulate and Iodine Samplers

To evaluate the CSM performance, heavy reliance was placed on comparison of the total iodine concentrations from the CSM with those obtained with Particulate and Iodine (P-I) Samplers. The P-I Samplers are "Maypack" type cartridges consisting of a series of filters and sorbent beds which collect particulate fission products and volatile iodine species. (A rack of P-I Samplers installed in the CDE vessel is shown in Figure 8.) The composition of the sorbent beds changed during their development program. This program is being conducted concurrently with the CSM development program. The current arrangement

Fig. 8 Cutaway view of CDE showing sampler locations.

of sorbent beds in the P-I Sampler, which is shown in Figure 9, appears to separate the iodine species satisfactorily. It consists of a glass fiber filter to collect particulates, a bed of cadmium iodide adsorbed on "Chromasorb-P" to collect elemental iodine vapors, a bed of copper zeolite to collect hypiodous acid vapors, a bed of silver zeolite to collect methyl iodide, and a bed of impregnated charcoal as a back-up trap. Although the early versions of the P-I Sampler did not separate the iodine species successfully, the sorbent beds retained all the iodine entering them. Consequently, the P-I Sampler was considered the reference total iodine sampler.

3.3 Raw and Filtered Gas Samplers

The Raw and Filtered Gas Samplers collected containment atmosphere samples for subsequent determination of xenon or methyl iodide concentrations by the chromatographic method described earlier. With the initial version of the Raw Gas Sampler, used in CDE Tracer Runs 1 through 4 and in the CDE runs with irradiated fuel pins, an atmosphere sample was obtained by drawing the sample through a 2-ft length of stainless steel tube into a gas sample cylinder. For subsequent runs, the Raw Gas Sampler was modified by adding a condenser upstream of the gas sample cylinder and lengthening the lead lines to 20 ft. This modified Raw Gas Sampler is shown pictorially in Figure 8. The Filtered Gas Sampler is similar to the Raw Gas Sampler except that a filter as well as the condenser were installed upstream of the gas sample bomb. There was no detectable loss of xenon or organic iodide as a result of adding the condenser, filter, and extra line length; the xenon or organic iodide concentrations in the gas samples taken with these three gas sampling systems were essentially identical. Consequently, the Raw and Filtered Gas Samplers are assumed to provide representative samples of noble gases and organic iodides in the CDE containment atmosphere.

3.4 CDE Particulate Tests

Cladding fumes vaporized from a heated, nonirradiated fuel pin also were used to test the early version of the CSM scrubber design (without the common discharge line). The particulates, primarily oxides of tin and uranium, present in the CSM scrubbed gas stream were collected on a

INC-A-15413

Fig. 9 Particulate and Iodine Sampler.

glass fiber filter. The tin content of particulate trapped on the filter was then measured by emission spectrometry (the uranium content was below the detection limit) and compared with the tin content of samples taken directly from the containment atmosphere and analyzed by the same method.

III. CSM PERFORMANCE TESTS

The CSM development program emphasized evaluation of the CSM performance for determining airborne noble gas, total iodine, and organic iodide concentrations, and for monitoring gross activity levels. The CSM was tested under simulated LOFT atmospheric conditions in the CDE facility with two xenon releases, three methyl iodide releases, and six "elemental" iodine releases. The CSM was also tested inconclusively in three early CDE tests with irradiated fuel pins. The reproducibility and precision of the CSM test results improved as the program progressed due to improvements in the CSM design and operating procedures, and improvements in techniques for sampling and analyzing the CSM effluent streams and the CDE containment atmosphere. The test results demonstrate that the CSM performance is strongly affected by operating parameters such as the gas sampling rate and the scrubbing solution composition. The results of the mixed fission-product and early elemental iodine tests in which the CSM operated with low gas sampling rates and scrubbing solutions without sodium thiosulfate were erratic and differed significantly from the results of the later tests. The results presented in the following sections primarily are from the later tests in which consistent results were obtained and in which the CSM was operated under conditions most similar to those recommended for LOFT. The presentation of results is divided into the following sections according to CSM objective or fission product form: noble gases, elemental iodine, methyl iodide, other iodine species, particulates, transient monitoring, and design evaluation.

1. NOBLE GAS SAMPLING

Two CDE tracer-level runs (Tracer Runs 1 and 2) were conducted with xenon-133 to determine the sampling accuracy of the CSM for xenon. The total concentration of xenon (including stable fission product xenon shipped with the xenon-133) injected into the CDE vessel was 0.01 volume parts per million. This concentration is higher than the fission-product xenon concentrations anticipated in the LOFT-ECCS tests

but represents only 10% of the normal atmospheric xenon concentrations^[4]. The airborne xenon-133 concentrations were constant throughout both runs. Less than 0.005% of the xenon injected was absorbed in the containment vessel condensate.

The xenon concentrations from the CSM agree closely with the values obtained from the Raw and Filtered Gas Samplers in both runs. The atmospheric xenon concentrations measured by the different samplers during CDE Tracer Run 2 are plotted in Figure 10. The results from Run 1 are similar. The total concentration of xenon measured by the CSM was determined from the xenon concentrations in the liquid and the gas samples. There is a 10 to 20% difference between results obtained by the two different methods of analysis--direct counting of the gas sampling cylinders, and chromatographic separation followed by trapping and counting of the xenon; however, when analyzed by the same method to eliminate the effect of systematic calibration errors, the CSM average xenon concentrations agree within 4% with those obtained by the other two samplers.

The xenon concentrations observed in the scrubbing solution, which ranged from 6 to 10% of the total xenon in the atmosphere samples taken by the CSM, agree reasonably well with those calculated from existing solubility data^[5]. This suggests that xenon and krypton solubility data can be used to estimate the effect of the subsequent increase in the gas sampling rate from 210 to 2000 ml/min on the fraction of xenon and krypton absorbed in the scrubbing solution. At this higher gas sampling rate, the fractions of xenon and krypton absorbed in the scrubbing solution should be negligible--about 1% of the total xenon and 0.5% of the total krypton in the sample stream.

2. TOTAL IODINE SAMPLING

The ability of the CSM to furnish total airborne iodine concentrations under simulated LOFT conditions was evaluated in six CDE runs in which an iodine sample consisting of primarily elemental iodine vapors mixed with small amounts of organic iodides and possibly hypoiodous acid was injected into the air-steam atmosphere in the CDE vessel. Following

Fig. 10 Xenon-133 concentration in CDE containment atmosphere during CDE Tracer Run 2.

injection, the vessel was cooled at a rate approximating that anticipated in LOFT. The initial total iodine (iodine-131 plus iodine-129 and stable iodine-127) concentrations in the 6 runs ranged from 0.054 to 7.1 $\mu\text{g}/\text{m}^3$. In most of the CDE runs, the CSM was operated and sampled for six hours after the iodine release. However, in Run 11 the CSM was operated for 3 hr, shut-down due to a mechanical failure, then restarted on the next day and operated for an additional 3 hr. The CSM operating conditions were summarized previously in Table I.

In each of the CDE elemental iodine runs, there was a rapid washout--40 sec to 3 min half-time--of 80 to 95% of the iodine initially injected. The iodine airborne after this initial washout consisted almost entirely of organic iodides and hypiodous acid vapors. The elemental iodine airborne immediately after injection was removed within 5 or 10 min of injection by the processes of absorption in the condensing steam and deposition on the walls. The organic iodides appeared to be formed as an impurity during preparation of the iodine injection source. The hypiodous acid was essentially all formed when the first gas samples were taken less than a minute after injection; however, it is not known whether it was formed before, during, or immediately after injection. Following the initial rapid washout, the total airborne iodine concentration decreased slowly over the rest of the run leaving from 5 to 15% of the iodine injected airborne at the end of the run.

The CSM total iodine concentrations are evaluated by comparison with values from the P-I Samplers. A measure of the consistency of the CSM values is also provided by comparison of the CSM values obtained at the two different gas sampling rates. The total airborne iodine concentrations from the P-I Samplers and CSM agreed at least to within 50% during the last four CDE elemental iodine runs. However, there were significant differences in the sampler performance in the different runs. In Runs 6 and 9 there was good agreement ($\pm 10\%$) between the total iodine concentration values of the CSM and P-I Samplers. This is shown in Figure 11 where the Run 6 total iodine concentrations are plotted. In Runs 10 and 11, significant discrepancies appeared between the total iodine concentration from the CSM at the two different gas sampling rates and between the CSM and P-I Sampler values. These are illustrated

Fig. 11 Iodine-131 concentration in CDE containment atmosphere during CDE Tracer Run 6.

by Figure 12 which shows the results of Run 11--the run in which the largest discrepancies occurred. The CSM values obtained at the two different gas sampling rates differed significantly in both halves of this run. In the last half of the run, both sets of CSM values were significantly higher than the P-I Sampler values. In the first half of the run, the CSM values at the low gas sampling rate were almost 50% higher than the P-I Sampler values. Likewise, in Run 10 (not shown) the CSM values at the two sampling rates differed by nearly 40%. In Run 10, the CSM values at the higher gas sampling rate agreed with those from the P-I Samplers.

The discrepancies in Runs 10 and 11 appear to be the result of experimental errors rather than fundamental problems with the CSM. There was definite evidence of irregularities in the CSM samples--particularly, the CSM gas stream samples--in these runs. Although the causes of the discrepancies have not been definitely determined, some possible causes are:

- (1) Failure of the CSM gas stream sampling packs: There is definite evidence that iodine penetrated the CSM gas sampling pack at the 2 liter/min gas sampling rate but not at the 1 liter/min gas flow. This could account for the discrepancy between the CSM values at different flow rates.
- (2) Calibration bias: The CSM and P-I Sampler concentrations are determined from the analysis of different types of samples and the measurement of different flow rates. A bias in the relative calibrations could account for differences between the CSM and P-I Sampler values.
- (3) Leak: A leak in the CSM gas lines could cause erroneous gas flow or volume measurements; however, no significant leak was observed in pre-run checks.

The discrepancies between the sampler values leave an uncertainty about the true iodine concentration values. In the absence of an absolute value for the airborne total iodine concentration, the true value

Fig. 12 Iodine-131 concentration in CDE containment atmosphere during CDE Tracer Run 11.

is assumed to be in the range bracketed by the sampler results. The results of the last four CDE runs then indicate that, at the worst, the CSM probably provides total iodine concentration values accurate to within $\pm 50\%$. Results comparable to those obtained in Run 6 (Figure 11) might be expected if the experimental errors could be eliminated.

The iodine aerosol in the CDE tests was not entirely representative of the fission-product aerosol anticipated in LOFT. Due to the rapid washout of elemental iodine, the airborne iodine in the CDE "elemental" iodine runs consisted almost entirely of organic iodides and hypiodous acid except for the first few minutes of the tests. Consequently, only the first two or three samples in each run have contained a significant fraction of elemental iodine. The CSM iodine concentrations obtained during the initial washout appear reasonable; however, it is difficult to compare samplers during this period of rapid change. The time at which a CSM sample was taken cannot be assigned precisely, because the length of time needed to draw the CSM sample streams through the CSM lines is not precisely known. Hence, the CSM performance on elemental iodine vapors can at best be considered only partially evaluated. Tests in a larger containment vessel in which elemental iodine is removed at a slower rate, for example, the Containment Systems Experiment (CSE) at BNWL, are needed to evaluate the CSM performance for elemental iodine vapors.

The mixture of iodine and other nuclides expected in LOFT will be more difficult to separate and analyze than the single nuclide (iodine-131) used in the CDE tracer tests. Also, the distribution of iodine forms released from a fuel pin could be different from those released in the tracer runs. As a result of difficulties discussed previously, the early CSM tests in the CDE fuel pin runs did not provide useful results. Hence, tests on a release of mixed fission products from an irradiated fuel pin are needed to confirm the CSM performance in the tracer tests.

The range of initial iodine concentrations in the CDE tests-- 0.054 to $7.1 \mu\text{g}/\text{m}^3$ --covers the most probable initial iodine concentrations in LOFT. However, the iodine concentrations anticipated for the cases of clad rupture of a single pin or complete core meltdown are outside the range covered in the CDE tests.

3. ORGANIC IODIDE SAMPLING

It would be advantageous for the CSM to separate the inorganic iodine species into the scrubbing solution stream leaving the organic species in the scrubbed gas stream. Unfortunately, solutions used to absorb inorganic iodides also absorb some organic iodides. Consequently, the organic iodide concentrations in the CSM scrubbed gas stream are lower than those in the containment vessel.

The absorption of a vapor into a scrubbing solution involves several steps. After contacting the solution, the vapor dissolves at the liquid surface, diffuses into the bulk of the liquid, and reacts with the reactive solutes. Factors which affect the absorption rate are the gas-liquid contact area, the solubility of the vapor, the reaction rate in the solution, and the gas-liquid contact time. These, in turn, are affected by the scrubber volume and the discharge line length and diameter (contact area and contact time), the scrubbing temperature (reaction rate and solubility), and operating parameters such as solution composition (reaction rate) and gas sampling rate (contact time).

Scrubbing solution composition and gas sampling rate are the operating variables affecting absorption which are most readily varied to improve the CSM performance. Hence, the effect of these operating parameters was evaluated in both laboratory tests and CDE runs with methyl iodide. In addition, some laboratory tests were conducted with ethyl iodide, because ethyl iodide was observed in the containment atmosphere during one CDE "elemental" iodine run. Basic thiosulfate mixtures were used in these tests because of their known ability^[8] to absorb elemental iodine. The operating parameters for the three CDE methyl iodide runs are listed in Table I.

The CSM scrubbing solution absorbed from 10 to 50% of the methyl iodide in the entering gas stream when operated at the recommended conditions: a 2 liter/min gas sampling rate and a scrubbing solution containing 1M sodium hydroxide and 0.007M sodium thiosulfate. The results of a typical CDE run are shown in Figure 13 where the ratio of the methyl iodide concentration in the CSM scrubbed gas samples,

Fig. 13 Relative methyl iodide concentration in the CSM scrubbed gas stream during CDE Tracer Run 8.

C, to the methyl iodide concentration in the containment atmosphere, C_A , (as determined by analysis of raw gas samples) is plotted against time after injection. The change in C/C_A during the run appears to be a result of the decrease in the containment (and the scrubbing solution) temperature during the run. Similar results were encountered in other tests with the same operating conditions. The methyl iodide loss due to absorption in the solution in these runs can be represented by an average correction factor, $C/C_A = 0.7 \pm 0.2$. Hence, it appears possible to obtain methyl iodide concentration values accurate to $\pm 30\%$ by dividing the results obtained by chromatographic analysis of CSM gas stream samples by the average correction factor, $C/C_A = 0.7$.

The scrubbing temperature, gas sampling rate, and solution composition all have important effects on sampling organic iodides. These effects are discussed in the following sections.

3.1 The Effect of Solution Composition

Organic iodides react with nucleophilic reagents--eg, thiosulfate and hydroxide ions--by a nucleophilic displacement-- S_N2 --mechanism^[7,9]. For example, methyl iodide reacts with the hydroxide ion:

The published^[7] reaction rate constants for the reaction of methyl iodide with thiosulfate and hydroxide ions, listed in Table II, show that the thiosulfate ion reacts much faster than the hydroxide ion.

TABLE II
REACTION RATE CONSTANTS OF METHYL IODIDE^[7]

Reactant	Bimolecular Reaction Rate Constant, liter/mole-sec	
	at 25°C	at 120°C (extrapolated)
Hydroxide ion	6.36×10^{-5}	0.55
Thiosulfate ion	0.0284	64
	0.0331	75

The results of the laboratory CSM scrubbing tests confirm that the thiosulfate ion reacts much faster than the hydroxide ion. In Figures 14 and 15, the fractions of methyl and ethyl iodide penetrating the scrubbing coil--ie, the fraction not scrubbed--are plotted against the concentration of thiosulfate or hydroxide ion in the scrubbing solution. As expected from the reaction rate constants, the thiosulfate solutions absorbed more methyl iodide than higher molarity hydroxide solutions; more methyl iodide passed through the 1M NaOH solution than the 0.01M $\text{Na}_2\text{S}_2\text{O}_3$ solution. It is apparent from Figures 14 and 15 that a large fraction of the methyl iodide will remain in the gas stream being sampled only if the thiosulfate ion concentration is low; however, hydroxide ion concentrations up to 1M can be used without excessive methyl iodide loss.

Figures 14 and 15 also show that ethyl iodide reacts much more slowly than methyl iodide with both thiosulfate and hydroxide ions. No reaction with the hydroxide ion and only a slight reaction with the thiosulfate ion was observed for ethyl iodide. The probable reason for the slower reaction rate of ethyl iodide is steric hinderance. The methyl group on the carbon atom to which the iodine is attached blocks the approach of the nucleophilic ion, resulting in the slower reaction rate. The slower reaction rate of ethyl iodide (and probably other heavy organic iodides) means that ethyl iodide will be easier to sample with the CSM than methyl iodide. (It also infers that containment sprays will be less effective for ethyl iodide than on methyl iodide.)

The best CSM methyl iodide sampling results--ie, highest C/C_A values--are obtained using the most dilute scrubbing solutions. However, some sodium thiosulfate and sodium hydroxide appears necessary for elemental iodine sampling. The solutions containing 1M sodium hydroxide and 0.007 to 0.01M sodium thiosulfate appear to provide a satisfactory compromise.

3.2 The Effect of Scrubbing Solution Temperature

The scrubbing solution temperature has opposing effects on solubility and reaction rate. While the solubility of methyl iodide decreases with increasing scrubbing solution temperature, the reaction

Fig. 14 The absorption of methyl and ethyl iodide by sodium thiosulfate solutions at 80°C.

Fig. 15. The absorption of methyl and ethyl iodide by sodium hydroxide solutions at 80°C.

rate increases with increasing temperature^[7]. Thus, an increase in scrubbing temperature could result in either an increase or decrease in scrubbing, depending on the relative importance of the solubility and the reaction rate.

The scrubbing temperature is the only variable affecting methyl iodide scrubbing which changed significantly during a given CDE run. The gas sampling rate was held constant at one of two rates, and the same scrubbing solution was used throughout the run; however, the temperature varied from about 110°C at the beginning of a run to ambient at the end of a run. Hence, it is reasonable to attribute the change in the relative methyl iodide concentration (C/C_A) in the CSM scrubbed gas stream samples during a run to the change in scrubbing temperature. This is illustrated in Figure 16 where the values, previously shown in Figure 13, of the fraction of methyl iodide in the CSM scrubbed gas stream, C/C_A , during CDE Run 8 are plotted against scrubbing temperature. Increasing the scrubbing temperature increased in methyl iodide absorption (lower C/C_A) apparently as a result of an increased reaction rate.

Figure 16 shows that the effect of the change in scrubbing temperature on C/C_A during a simulated LOFT temperature transient is moderate when scrubbing 2 liters/min of gas sample with a basic solution of dilute (0.005M) sodium thiosulfate. When the fraction of the methyl iodide absorbed is small, changes in the absorption rate do not produce large variations in the relative concentration of methyl iodide in the scrubbed gas stream. For example, when C/C_A is 0.85, a 100% increase in the fraction absorbed (from 0.15 to 0.30) results in only an 18% reduction in C/C_A (from 0.85 to 0.70).

3.3 Effect of Gas Sampling Rate

The time the atmosphere sample is in contact with the scrubbing solution depends on the length (or volume) of the common discharge line leading from the scrubber to the separator and on the rate at which the gas flows through the scrubber and discharge lines. The length of the discharge lines of the LOFT CSM's will depend on the position of the scrubbers in the LOFT building and the routing of the discharge lines-- a variable which is dictated by the building design and sampling objectives.

Fig. 16 Effect of scrubbing solution temperature on methyl iodide scrubbing.

Thus, control of the gas sampling rate is the only means by which the gas-liquid contact time can be varied.

In the CDE methyl iodide runs, an increase in the gas sampling rate resulted in an increase in the methyl iodide concentration in the CSM scrubbed gas stream, C/C_A , because of the reduced contact time.

The exponential expression:

$$\frac{C}{C_A} = \exp - \frac{K}{Q_G} \quad (1)$$

where:

C is the concentration in the scrubbed gas,

C_A is the concentration in the containment vessel,

Q_G is the gas flow rate,

and K is a constant which depends on the scrubbing variables, also predicts an increase in C/C_A with increasing gas flow rate. This exponential equation can be derived theoretically by assuming that the absorption rate is proportional to the methyl iodide concentration in the gas stream and the contact time. Table III shows that Equation (1) correlates the effect of increasing the gas flow quite well.

TABLE III
COMPARISON OF EXPERIMENTAL AND CALCULATED EFFECT OF DOUBLING GAS
SAMPLING RATE ON RELATIVE METHYL IODIDE CONCENTRATION IN CSM
SCRUBBED GAS STREAM

CDE Run	Measured C/C_A With 1 liter/min Flow	C/C_A With 2 liter/min Flow	
		Calculated Extrapolation	Measured
7	0.46	0.68	0.69
7	0.50	0.71	0.71
8-conc. sol'n.	0.40	0.63	0.75
8-conc. sol'n.	0.34	0.58	0.58
8 dil. sol'n.	0.72	0.85	0.81
8 dil. sol'n.	0.63	0.79	0.75

Column 2 lists C/C_A values measured at a 1 liter/min gas sampling rate. These values were extrapolated using Equation (1) to a 2 liter/min gas sampling rate and the extrapolated values listed in Column 3. For comparison, measured values of C/C_A obtained at a 2 liter/min gas sampling rate with the same scrubbing temperature and solution as used to obtain the values in Column 2 are listed in Column 4. The agreement between Columns 3 and 4, which is well within the accuracy of the data, shows Equation (1) provides a good means of estimating the effect of changing gas flow on methyl iodide sampling.

Equation (1) suggests that C/C_A can be increased further by increases in the gas sampling rate. However, a gas sampling rate of 2 liters/min is recommended for use in LOFT because most of the tests have been conducted at this flow rate. Operation at higher flow rates could lead to unforeseen difficulties.

4. HYPOIODOUS ACID SAMPLING

The separation of inorganic iodides from the organic iodides by CSM was complicated by the presence in the CDE containment atmosphere of a volatile, unreactive, iodine species which has tentatively been identified^[10] as hypiodous acid. The P-I Sampler uses a bed of copper zeolite (in sulfide form) to trap hypiodous acid and separate it from the organic iodides which then flow to a subsequent bed (the elemental iodine is removed before it reaches the copper zeolite bed). Hypiodous acid, as determined by the P-I Samplers, usually comprised nearly half of the iodine airborne in the CDE containment atmosphere following the initial washout of the elemental iodine (the balance of the airborne iodine was mostly methyl iodide).

Hypiodous acid and organic iodides were determined to be the major components of the CSM scrubbed gas stream by sampling with P-I type sorbent beds. A comparison of the hypiodous acid concentrations in the containment vessel and in the CSM scrubbed gas indicates that about half of this iodine form was scrubbed by the CSM scrubber. The relative effects of solubility and chemical reaction cannot be determined from this data. Preliminary laboratory tests on scrubbing an iodine vapor

believed to be hypiodous acid indicate that this species is a little more soluble than methyl iodide but that it does not react appreciably with thiosulfate solutions or bases.

The difficulty of separating hypiodous acid from organic iodides does not affect the total iodine concentration values from the CSM as long as the total iodine concentrations in both the gas and liquid streams are measured. However, two gas stream samples must be taken--one for total iodine and one for organic iodides--as well as a liquid stream sample, to determine both total iodine and organic iodide concentrations with the CSM.

5. PARTICULATE SAMPLING

Particulates are of importance for the CSM primarily because the iodine adsorbed on them constitutes a part of the total airborne iodine. The CSM is not intended to sample particulates per se because the SP-I Sampler will be used to determine the particulate concentrations in LOFT. Particulates are not expected to be a major fission-product form in the LOFT-ECCS tests where clad failure without fuel melting is expected. However, there are conceivable cases--eg, gross core meltdown--in which particulates could be important. Unfortunately, enough data are not available on the adsorption of low concentration iodine on particulates or on particulate releases under LOFT-ECCS conditions to assess the importance of particulates. Hence, the necessity of determining the performance of the CSM on particulates should be evaluated.

The scrubbing efficiency of the CSM scrubber for particulates depends on the particle size. The scrubber is designed to wash particulates into the liquid stream. In laboratory tests (which are discussed in detail in Section 5.1) with fluorescein dust, an early version of the scrubber effectively scrubbed particles larger than 0.2 micron diam, but most of the finer particles remained in the scrubber gas stream. CDE test results likewise indicated effective scrubbing of the larger particles; however, the finer particles airborne immediately after the gross meltdown of a Zircaloy-clad fuel pin penetrated the scrubber in significant quantities.

Changes in the CSM design and operating conditions since these early tests have probably improved the scrubbing performance of the CSM for particulates. The addition of the common discharge line and cooler through which the atmosphere sample and scrubbing solution flow provides additional contact of the scrubbing solution and gas. Condensation of steam in this common discharge line will wash particulates into the scrubbing solution by the processes of diffusophoresis, and nucleation followed by settling. Conversely, the increase in gas sampling rate after the fluorescein tests may reduce scrubbing by reducing the contact time in the scrubber; however, the addition of the common discharge line is expected to have the greater effect.

Many particulate species such as UO_2 , SnO_2 , Mo, and TeO_2 cannot be sampled representatively with the CSM, because they are insoluble in the CSM scrubbing solutions. Insoluble particulates tend to settle out in the long lines and, thus, are lost. The CSM scrubbing solutions do, however, dissolve iodine adsorbed on the particulates.

The CSM scrubber is not an isokinetic sampler. The three sampling ports in the vertical wall of the scrubber are oriented perpendicular to, rather than parallel with, the vertical convective flow of the containment-atmosphere anticipated in the LOFT. Isokinetic sampling is not required^[11] for the accurate sampling of gases and particulates smaller than 5 microns; these small particles essentially have no inertia. Particles released in fuel-meltdown experiments conducted in the CDE^[3] and at ORNL^[12] have been significantly smaller than 5 microns. Hence, the CSM appears capable of obtaining a representative sample of the particulates anticipated in LOFT, without isokinetic sampling.

The laboratory fluorescein scrubbing tests and the CDE particulate tests are discussed in detail in the following sections.

5.1 Fluorescein Tests

The results of the fluorescein scrubbing tests (described in Section II-2.2) showed that changes in the gas sampling rate (in the range from 160 to 650 ml/min) and changes in spray nozzle had no significant effect on scrubbing efficiency. The experimental scrubbing efficiencies for fluorescein dust varied from a low of 95.3 to a high of 99.5%; however, most efficiencies were between 98 and 99%.

The published literature on the impaction of particles on moving spray drops offers an explanation^[13] for the observed lack of effect of the gas sampling rate and spray nozzle on the scrubbing efficiency. The impaction efficiency for spheres, ie, the ratio of the effective cross section to the actual cross section, is a function of $\sqrt{\Psi}$,

where:

$$\sqrt{\Psi} = D_p \sqrt{C_p V / 18 \mu D_c} \quad (2)$$

D_p = diameter of the particle

C = Cunningham slip correction

ρ_p = density of the particle

V = relative velocity between particle and collector

μ = viscosity of the gas

D_c = diameter of the collector

The impaction efficiency, η , which is shown in Figure 17 as a function of $\sqrt{\Psi}$, rises sharply as $\sqrt{\Psi}$ increases from 0.15 to 1. Outside of this narrow range, changes in $\sqrt{\Psi}$ have little effect.

Fig. 17 Impaction efficiencies of spherical collectors^[13].

Impaction efficiencies, taken from the curve of Ranz and Wong^[13], for the conditions* of the spray scrubbing tests are tabulated in Table IV for various drop and particle sizes. The impaction efficiencies in Table IV are low for particles smaller than 0.2 micron but appreciable for larger particles. The particle size distribution in Figure 7 shows that 98% of the fluorescein mass is contained by particles larger than 0.2-micron diam. This 98% fraction agrees with the measured scrubbing efficiencies. It thus appears that the cutoff of impaction efficiency with decreasing particle size is the effect which controls the measured scrubbing efficiencies. Under the conditions of the experiments, the scrubber removes all the dust larger than 0.25 micron, a varying fraction of the dust between 0.15- and 0.25-micron diam, and very little of the dust smaller than 0.15-micron diam. In more general terms, the cutoff conditions of the spray scrubber are an impaction efficiency of 0.2 or a value of $\sqrt{\Psi}$, as defined in Equation (2), of 1/3.

TABLE IV
IMPACTION EFFICIENCIES OF SPRAY DROPS

Particle Diameter, Micron	0.6	0.4	0.3	0.2	0.15	0.1
70 micron droplet size	0.8	0.6	0.4	0.15	0.02	0.0
50 micron droplet size	0.83	0.73	0.55	0.22	0.05	0.0

5.2 CDE Tests

In the CDE tests in which fuel pins were melted^[3], particulates consisting primarily of tin fumes from the Zircaloy cladding were released from the molten fuel pins. Particulate concentrations of 1 to 50 mg/m³ were measured in the containment atmosphere shortly after meltdown^[3]. The individual particles ranged in size from 0.001 to 0.7 microns and agglomerated into clumps with a diameter of around 1 micron at the time of sampling--usually 15 min after meltdown.

The CSM was operated shortly after meltdown during two CDE runs

*V = 190 ft/sec (70% of the velocity from a frictionless nozzle at 500 psi), $\rho_p = 0.8$ g/ml, C = 1.

with irradiated fuel-pin meltdowns. In these tests, large quantities of iodine and particulate fission products, eg, tellurium, were found in CSM gas-stream samples and plated-out in the CSM gas line. In another CDE run in which an irradiated fuel pin was melted, the CSM was not operated until two hours after the meltdown. In this run, no particulate fission products and very little iodine were found in the scrubbed gas stream.

The CSM scrubbing efficiency for particulates was also evaluated in a "cold" CDE test with a tin aerosol generated by heating an unirradiated fuel pin. Beginning 25 min after meltdown, samples of the particulates in the scrubbed CSM gas stream were collected on filters and analyzed by emission spectrometry. Comparison of the quantities of tin collected on the filters showed that 95% (on a mass basis) or more of the tin particles were removed by the CSM scrubber.

It appears that only the fine particulates penetrated the CSM scrubber during the CDE (fuel pin) runs. Shortly after meltdown, a significant quantity of particles finer than 0.1 micron were airborne in the containment atmosphere. These fine particles probably carried adsorbed fission products through the scrubber during the early part of the two "hot" runs. Late in the CDE runs, little penetration of particulates through the scrubber occurred because the fine particles agglomerated into larger clusters which were readily scrubbed.

6. GROSS ACTIVITY MONITORING

Gross-activity monitors are located on both the scrubbing-solution and scrubbed-gas streams to monitor the gross-activity levels. These monitors should determine the time of the arrival of fission products in the LOFT containment atmosphere and continuously record gross-activity levels in the containment atmosphere.

The use of gross-activity monitors on the LOFT CSM gas and liquid streams is complicated by the time lag and backmixing which occur in drawing the sample streams through the long sample lines. The CSM response to an activity change depends on the gas and liquid flow rates, the line lengths and volumes, and possibly, the system geometry. In

general, shorter time lags and less backmixing can be expected if the gas and liquid flow rates are increased or if the line lengths and volumes are reduced.

The response of the CSM gross-activity monitors to the iodine injections in the CDE tracer runs was used to evaluate the CSM response to sudden activity releases. The line length used during recent CDE runs--about 130 ft from the scrubber to the separator--is typical of line lengths expected in LOFT. The brief releases--1/2 to 2 minutes duration--used in the CDE tests were assumed to provide an instantaneously mixed containment atmosphere. Hence, any time lag between injection and the first response of the gross-activity monitors was attributed to the CSM; any deviation from a step function response was attributed to back-mixing in the scrubber and discharge lines.

Typical responses of the gross-activity monitors to an iodine release in CDE are shown in Figures 18 and 19 which are tracings of the recorder charts from liquid and gas stream gross-activity monitors, respectively, from CDE Run 10. (The gaps in Figure 19 were a result of gas samples being taken upstream of the gross-activity monitor.) The liquid-stream, gross-activity monitor showed a 5-min initial time lag while the iodine traveled through the CSM lines followed by a rapid rise (20 sec time constant) to the peak activity reading. The activity reading remained at the peak level for 2 minutes, then dropped a factor of 5 with a 2-min half-time as it followed the iodine washout in the containment atmosphere. For the rest of the run, the liquid-stream gross-activity reading decreased slowly to a nearly constant value.

The gas-stream, gross-activity monitor showed a shorter initial time lag--3-1/4 min--and a slower initial rise in reading--1-1/2 min time constant--than the liquid stream. Following the peak, the monitor reading dropped for a few minutes, then became essentially constant for the rest of the run.

It is difficult to evaluate the responses of the gross-activity monitors without detailed knowledge of the actual activity concentrations during the CDE runs. The liquid-stream monitor traces agreed closely with the gas and liquid sample analyses, and the shape of the

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Fig. 18 Response of the CSM liquid stream gross activity monitor to an iodine release--CDE Tracer Run 10.

Fig. 19 Response of the CSM gas stream gross activity monitor to an iodine release--CDE Tracer Run 10.

liquid-stream monitor trace conforms to the shape expected for a sudden iodine injection except for the flat peak (a sharp peak was expected). From Figures 18 and 19, it appears that by correcting for a predetermined time lag, the time of an instantaneous release could be determined within 2 or 3 min.

There was no detectable deposition of iodine in the liquid-stream monitor in which the scrubbing solution flowed through a "Kynar" pipe section. In the earlier runs with fission products released from irradiated fuel pins, there was significant deposition of insoluble fission products--tellurium, molybdenum--in the liquid-stream monitor; however, this was eliminated by placing a filter upstream of the monitor to remove particulates. Deposition of iodine in the aluminum* coils of the gas-stream, gross-activity monitor produced a significant increase in background activity during the elemental iodine runs. At the end of these runs, the background radiation contributed over half of the total monitor reading. Efforts to decontaminate the gas-stream monitor with decontamination solution ("Kern 85D" acidified to pH 2) at 70°C were unsuccessful.

The detectors used in the gross-activity monitors during the CDE runs were different than the detectors which will probably be used in LOFT. A set of 6 G-M tubes wired in parallel was used in most of the CDE runs because of the low activity levels. While the G-M tubes were satisfactory for the CDE runs in which a known quantity of single nuclide was released, detectors, such as an ion chamber, with a wider range and a response which is less dependent on gamma-ray energy^[14] than a G-M tube would probably be better for the LOFT. The possible fission-product concentrations in LOFT range over several orders of magnitude, and the activity will be a mixture of nuclides of different gamma-ray energies.

The problems of calibrating the LOFT gross-activity monitors and interpreting their readings have not been resolved. Both CSM effluent streams will contain mixtures of nuclides with different gamma-ray energies and branching ratios. The ratios of nuclides will change during the test as the short-lived nuclides decay. The conversion of the

*Aluminum was the only material with sufficient ductility to be bent in the desired coil radius.

monitor readings to gross activity concentrations in $\mu\text{Ci/liter}$ will require extensive calibration along with estimates of the proportions of various nuclides. Interpretation of the monitor readings may also present uncertainties. The gross-activity monitors will respond primarily to the most active fission products which will be the noble gases--eg, krypton-88, xenon-138--in the gas stream and their daughters--rubidium-88, cesium-138--in the scrubbing solution stream. If most of the iodine released from the fuel is retained in the core by the coolant as is expected, the radio-iodine in the containment atmosphere will be a relatively minor--<10%--contributor to the gross activity levels.

7. DESIGN EVALUATION

The design of several CSM components should be re-evaluated before finalizing the CSM design for LOFT. Possible problems and potential improvements are discussed in this section.

Several different types of gas pumps have been used during the development program; all have caused problems. The carbon vane pump which failed to operate during Run 11 had previously operated flawlessly for three CDE runs and many cold tests. The use of spare pumps connected in parallel and the development of maintenance procedures for the gas pumps are recommended for LOFT..

The liquid control system failed during CDE Run 11. This system pumps the scrubbing solution from the separator at a controlled rate to prevent the solution from flooding the gas line while maintaining a liquid seal in the separator to prevent the gas from going into the liquid line. When gas sampling flow was increased from a low to a high rate, scrubbing solution in the line leading to the separator was swept into the separator by the increased gas flow at a rate greater than the liquid pump could remove. The separator then overflowed and flooded the gas line. Although the CSM gas sampling rate probably will not be changed during the LOFT, the LOFT blowdown pressure surge might also cause a sudden surge of scrubbing solution into the separator. Hence, an overflow prevention system should be added to the LOFT CSM to prevent any overflow from the separator into the gas line.

The tubing pump used to regulate liquid flow from the separator worked satisfactorily in the CDE runs in which the maximum pressure was 22 psig. However, at pressures greater than 25 psig, this pump cannot maintain the liquid seal in the separator. If the CSM is operated at containment pressures greater than 25 psig, a pressure regulator could possibly be placed upstream of the pump to regulate the inlet pressure.

The need for re-evaluation of the CSM gross gamma monitor design and the decontamination procedures for these monitors is discussed in Section III-6.

The liquid and gas stream total iodine sampling systems should also be re-evaluated. The liquid samples were collected in 15 ml glass bottles capped with a rubber diaphragm. To facilitate the counting of low-level samples, the use of a larger sample container with a better counting geometry should be considered. If the 15 ml glass bottles are used, the absorption of iodine in the rubber diaphragm, which occurred in some tests, should be studied in solutions with low sodium thiosulfate concentrations. The absorber packs used to sample the total iodine concentration in the scrubbed gas stream should be redesigned to eliminate the iodine penetration observed in some of the CDE runs at the 2 or 3 liter/min gas sampling rate. The use of a flow totalizer to measure the volume of the gas sample going through the gas sampler is recommended.

Leaks were troublesome throughout the CSM development program. The LOFT CSM should be designed to facilitate leak checking. The number of connections should be minimized. It is also necessary to locate the discharge line from the scrubber to the separator with a downward or level slope to allow drainage of the solution.

IV. CONCLUSIONS AND RECOMMENDATIONS

A Continuous Sampler-Monitor was developed to determine the fission-product concentrations in the LOFT containment-building atmosphere. A prototype CSM was tested in the 86-ft³ CDE vessel under atmospheric conditions simulating those anticipated in LOFT with releases of xenon, methyl iodide, and an iodine mixture consisting initially of mostly elemental iodine mixed with small amounts of organic iodides and hypoiodous acid.

The performance of the CSM in the tests with a single isotope is summarized and recommendations for further work are given in the following paragraphs.

1. CONCLUSIONS

Noble Gas Sampling

The noble gases, xenon and krypton, present few sampling problems. Except for a small fraction--expected to be less than 1%--which dissolves in the scrubbing solution, the noble gases remain in the CSM scrubbed gas stream where their concentrations can be determined by analysis of gas stream samples. In CDE tests with xenon-133, the xenon concentrations determined from CSM samples and from gas samples taken directly from the containment atmosphere agreed within 4%.

Total Iodine Sampling

The CSM provides total iodine concentrations accurate to within 50%. In two CDE runs, the CSM and P-I Sampler total iodine concentrations agreed within 10%; however, in two later runs, there was a divergence probably due to experimental errors between the values obtained by the CSM at different gas sampling rates and between the CSM and P-I Sampler values. The largest difference observed between the CSM and P-I Sampler results was less than 50%.

When the airborne iodine consists primarily of elemental iodine, most of the iodine will be present in the CSM liquid stream samples; however, after the elemental iodine washes out, most of the iodine will

consist of organic iodides and hypiodous acid, most of which will be present in the gas stream samples. Hence, the iodine content of the gas stream, as well as that of the liquid stream, must be determined to obtain reliable total iodine concentrations over the entire run. The gas stream total iodine sample is in addition to the organic iodide sample.

Organic Iodide Sampling

The organic iodide concentrations in the containment atmosphere were determined by chromatographic separation of the organic iodide in samples taken from the CSM scrubbed gas stream, followed by analysis by gamma-ray spectrometry. This value was then corrected for the organic iodides absorbed by the scrubbing solution. In a given run with a constant gas flow and solution composition, the fraction of organic iodide absorbed by the scrubbing solution varied as a result of the variation in containment temperature. When the CSM was operated at the recommended conditions--a gas sampling rate of 2 liters/min and a scrubbing solution containing 1M sodium hydroxide and 0.007M sodium thiosulfate--the fraction of methyl iodide absorbed by the scrubbing solution was relatively constant. The CSM then provided organic iodine concentrations accurate to $\pm 30\%$ using the average absorption correction.

Gross Activity Monitoring

The gross activity monitors on the CSM gas and liquid streams provided gross activity traces in CDE elemental and methyl iodide runs which agreed with the behavior expected in these runs. There was a time lag of around 5 min between the time of sampling and the time the activity was detected by the monitors due to the time required to transport the sample through the 130-ft line which simulated the length needed for the LOFT installation. The activity traces obtained in elemental iodine and methyl iodide tests indicate that by correcting for a time lag of about 5 min, the time of a sudden activity release can be determined with an accuracy of 2 or 3 min with the CSM gross activity monitors.

2. RECOMMENDATIONS

Reliable CSM performance should be demonstrated for a fission-product mixture, including short-lived nuclides and particulates, released from an irradiated fuel pin under LOFT-ECC conditions. The fission product aerosol in the LOFT containment atmosphere will contain many nuclides in several different chemical forms, possibly mixed with nonactive particulates. This fission product mixture will be more difficult to separate and analyze than the single isotopes used in the CDE tracer runs, and the distribution of iodine species may differ from that released in the "elemental" iodine tracer runs. Also, possible precursor effects from short-lived nuclides--eg, formation of cesium-138 particles from gaseous xenon-138 and iodine-132 vapors from particulate tellurium-132--should also be investigated.

The CSM should be tested in a large containment vessel. Due to the rapid washout of elemental iodine in the 86-ft³ CDE vessel, the airborne iodine during most of each CDE run consisted essentially of organic iodides and hypiodous acid. In a large vessel, elemental iodine concentrations would remain at a high level long enough to permit performance of the CSM on elemental iodine to be evaluated.

A fundamental study should be conducted to evaluate iodine behavior at low concentrations. The mechanisms and kinetics of organic iodide and hypiodous acid formation are poorly understood. The formation of organic iodides and hypiodous acid may be affected by the conditions of the iodine release--eg, temperature, humidity, moisture presence, and coolant composition. Hence, tests with irradiated fuel pins under LOFT-ECC conditions and a knowledge of formation mechanisms are needed to predict the relative concentrations of the different volatile iodine forms in LOFT. The possibility of organic iodides other than methyl iodide should be considered; ethyl iodide was observed in one of the CDE "elemental" iodine runs.

The selection of optimum CSM operating parameters involves a compromise between the requirements of scrubbing inorganic iodine forms and not scrubbing organic iodides. The following operating parameters

provided satisfactory CSM performance in CDE tests and are recommended for use in LOFT:

Gas sampling rate	2 standard liters/min
Scrubbing solution composition	<u>1M</u> sodium hydroxide 0.007 <u>M</u> sodium thiosulfate
Scrubbing solution flow	110 ml/min

Although the CSM performed satisfactorily in most of the CDE runs, some of the components have caused problems or appear inadequate for the LOFT Integral Test Program. Hence, a careful re-evaluation and possible redesign of the gross-gamma monitors, gas pump, liquid discharge metering pump, liquid discharge control system, liquid samplers, and gas stream samplers are recommended. Potential modifications are discussed in Section III-7.

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VI. APPENDIX

CSM DESIGN DETAILS

The CSM concept and general design are presented in Section II-1 of this report. This appendix presents a more detailed description of the CSM tested in the CDE facility. Figure A-1 shows a detailed flowsheet of the CSM installation and Figure A-2, the design of the CSM scrubber.

Scrubbing solution is drawn from a storage tank by a piston pump, then pumped through a pressure tank (Lapp Pulsatrol Model 38) which dampens pressure surges. The scrubbing solution flow rate depends on the pressure of the solution being supplied to the spray nozzle (shown in Figure A-2) which acts as a metering orifice. The solution feed pressure is controlled by the pressure relief valve, CSM-9. The pump is operated at excess capacity, and the excess solution returns to the tank through the pressure relief valve. The scrubbing solution is filtered through a sintered metal filter (10- to 15-micron pore size) to prevent particles from clogging the fine spray nozzle. Before reaching the scrubber, the solution passes through a 100-ft warming coil (which simulates the anticipated LOFT line length) in the CDE vessel in which the solution warms to the vessel temperature.

The scrubber (shown in Figure A-2) consists of a 1-1/2-in. ID by 12-in. long "Teflon" TFE cylinder into which the scrubbing solution is sprayed downward from the top. The atmosphere sample enters through three sample inlets in the side located a little above the spray sheet, then flows down through the spray and discharges with the scrubbing solution through a common discharge line.

It is important that leakage into or out of the scrubber be avoided. Leakage of moisture which condenses on the outside of the scrubber into the scrubber introduces extraneous activity and results in unrepresentative samples. Leakage of scrubbing solution could, in a small vessel, change the composition of the vessel condensate and perturb the iodine behavior in the vessel. The large cap serves as an "umbrella" to prevent condensate from running into the sampling inlets. The rest of the

A-1 CDE Continuous Sampler-Monitor flowsheet.

[illegible]

SCRUBBER BODY - ITEM 1

SECTION 13-13

CAP - ITEM 2

SECTION A-A

A-2 Continuous Sampler-Monitor spray scrubber.

scrubber was machined from a single piece of TFE to avoid unnecessary joints; however, there is still a potential for leakage at the connectors located at the top and bottom.

A 100-ft delay coil is added to the 3/8-in. ss discharge line (making 130 ft total length) from the scrubber to simulate the line lengths anticipated in LOFT. Downstream of the delay coil, the gas and liquid effluents pass through a heat exchanger in which they are cooled below ambient temperature to reduce the water vapor content of the gas and prevent condensation further downstream in the gas line. The gas and liquid effluent streams are separated by gravity in the separator which consists of a 1-1/2-in. pyrex pipe tee.

The liquid level in the separator is controlled by a system consisting of a differential pressure transmitter, a controller, and a variable speed metering pump to prevent liquid from overflowing into the gas line or air from going down the liquid line. The D/P transmitter measures the difference in liquid level between the separator and the reference pot (which drains through valve CSM-67). Some problems were encountered with air pockets in the lead lines to the D/P transmitter. The D/P transmitter and lead lines must be located below the reference liquid level and carefully purged with water to remove all air pockets. The metering pump which pumps (or throttles) the flow of solution from the separator is a Masterflex* tubing pump with a variable speed motor. With "Viton" rubber tubing, the tubing pump can regulate the liquid discharge from the scrubber at containment pressures up to about 25 psig. At containment pressures greater than 25 psig, leakage through the tubing pump becomes excessive, and air is blown through the pump into the liquid line. If the CSM is operated at containment pressures greater than 25 psig, a pressure regulator could be used upstream of the tubing pump to regulate the inlet pressure. The control signal (output) from the controller is amplified by a special motor speed control circuit (circuit diagram shown in drawing 036-0650-10-400-400415) to power the metering pump.

*Cole-Parmer Instrument Co., Chicago.

Liquid samples are drawn from the liquid line downstream of the metering pump through a short length of 1/8-in. ss tubing. The samples were collected in 15-ml glass bottles capped with a rubber diaphragm. The sample was injected into and flushed through the bottles with a pair of hypodermic needles which punched through the diaphragm. The liquid sample entered the bottle through one needle and overflowed through the other. Two bottles were taken in sequence during each sampling period; the first one was used to flush the sampler and the second, for the sample. A positive pressure was maintained at the liquid sampler by placing a check valve (CSM-88) at the downstream end of the liquid line to create a back pressure.

Prior to entering the liquid stream gross-activity monitor, the solution was filtered to collect particulate fission products and prevent their settling inside the gross-gamma monitor and raising its background. The liquid stream gross-gamma monitor (labeled liquid radiation monitor) used G-M tubes (except for CDE Run 11 where a NaI(Tl) scintillator was used) to monitor the activity flowing through a pipe inside a lead cave. The output from the G-M tubes was averaged by a count-rate meter whose output was recorded. After going through the monitor, the liquid stream was discharged to the hot waste system.

The gas stream leaving the separator was sampled with two sets of samplers. Gas samplers HC-1-603 through 606 were 10-ml charcoal packs designed to collect all iodine forms in the gas being sampled. The gas sample bombs (150 ml) were used to collect gas samples which were analyzed for organic iodide concentrations by chromatographic techniques.

The gas stream gross-activity monitor (labeled gas radiation monitor) was similar to the liquid stream monitor except for the design of the coil in which the gas stream activity was monitored. Downstream of the monitor, the gas stream was cleaned by charcoal packs, a drier, and a filter. Next, a thermal mass flowmeter measured and recorded the gas flow rate.

The gas flow rate was controlled by a D/P flow controller--Moore* Model 63-BD--and a pair of needle valves connected in parallel. The

*Moore Products Co., Spring House, Pennsylvania.

The D/P flow controller maintained a constant pressure difference across the needle valves regardless of the upstream pressure. When a constant pressure was maintained downstream of the needle valves, the flow rate depended only on the valve opening. The use of two (only one is needed) needle valves permitted changes in flow rate. A vacuum pump (HC-1-202) provided a positive pressure difference when the containment vessel pressure is low. The pressure downstream of the flow control needle valves was regulated by a vacuum relief valve--CSM-19. The exhaust from the vacuum pump went to the off-gas system.