MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0059843 1

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

Cy 2

ORNL-4253
UC-80 – Reactor Technology

REMOVAL OF IODINE VAPOR FROM AIR AND STEAM-AIR ATMOSPHERES IN THE NUCLEAR SAFETY PILOT PLANT BY USE OF SPRAYS

L. F. Parsly
J. K. Franzreb

OAK RIDGE NATIONAL LABORATORY
CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.
UCN-7969
(3 3-67)

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

ORNL-4253

Contract No. W-7405-eng-26

REACTOR DIVISION

REMOVAL OF IODINE VAPOR FROM AIR AND STEAM-AIR ATMOSPHERES IN THE NUCLEAR SAFETY PILOT PLANT BY USE OF SPRAYS

L. F. Parsly
J. K. Franzreb

JUNE 1968

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES

3 4456 0059843 1

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

CONTENTS

Abstract 1

Introduction 1

Experimental Equipment and Procedures 3

Interpretation of Gamma Recorder Data 6

Experimental Results 6

Analyses of Data 8

Discussion of Experiments 11

Effect of Solution Composition 11

Effect of Condensation 12

Comparison with Analytical Model 12

Summary and Conclusions 13

REMOVAL OF IODINE VAPOR FROM AIR AND STEAM-AIR ATMOSPHERES IN THE NUCLEAR SAFETY PILOT PLANT BY USE OF SPRAYS

L. F. Parsly J. K. Franzreb

ABSTRACT

This report presents the results of a series of nine experiments in which iodine vapor was released into the model containment vessel of the Nuclear Safety Pilot Plant and then removed by sprays. The experiments show that a half-life for iodine vapor removal of less than 1 min can be achieved using a type of nozzle proposed for installation in power reactor containment buildings at liquid mass flows currently being proposed. Thus they show that spray systems can be designed which are capable of removing iodine from containment building atmospheres rapidly enough to make a significant reduction in the 2-hr dose due to iodine leakage.

Little effect of solution composition on spray performance was found; condensation of steam on the drops also had no significant effect.

Fair agreement was found between the experimental results and a proposed analytical model.

INTRODUCTION

During the past year, several reactor designs have been proposed[1-4] in which containment sprays are to be used to remove both heat and airborne fission products from the containment atmosphere in the unlikely event of a loss-of-coolant accident. To develop data to substantiate the reduction of airborne activity to acceptable levels by the sprays, the U.S. Atomic Energy Commission initiated a spray and absorption technology program.[5] Since the Nuclear Safety Pilot Plant was designed to run spray tests under simulated accident conditions as well as to do fission product release and transport studies, it was convenient to do pilot-plant-scale tests of spray performance as one part of the overall program. The NSPP vessel is large enough so that spray nozzles suitable for large containment buildings can be tested in it.

Preliminary evaluation of the problem indicated that it could be broken down into three parts: (1) removal of iodine vapor (and HI), (2) removal of methyl iodide, and (3) removal of particles. The general behavior on each of these is summed up as follows:

1. Iodine vapor is moderately soluble in water[6] (distribution coefficients at room temperature are reported to be of the order of 100), and significant liquid film as well as gas film resistance

[1]Consolidated Edison Co. of New York, Inc., Indian Point Nuclear Generating Unit No. 3, Preliminary Safety Analysis Report, vols. I and II, Parts A and B, Docket 50-286, April 1967.

[2]Zion Station Preliminary Safety Analysis Report, Commonwealth Edison Co., vols. I and II, Docket 50-295, June 1967.

[3]Metropolitan Edison Co., Three Mile Island Nuclear Station Unit 1, Preliminary Safety Analysis Report, vols. I, II, and III, Docket 50-289, May 1967.

[4]Nuclear Plant, Diablo Canyon Site, Pacific Gas and Electric Co., Preliminary Safety Analysis Report, vols. I and II, Docket 50-275, January 1967.

[5]T. H. Row, "Spray Technology Program," ORNL-TM-2186 (to be published).

[6]R. F. Taylor, "The Absorption of Iodine Vapor by Aqueous Solutions," *Chem. Eng. Sci.* **10**, 68 (1959).

would be expected. However, iodine reacts very rapidly with reagents such as sodium thiosulfate and sodium hydroxide, and the liquid film resistance can be eliminated by using a sufficient concentration of these reagents.[7,8] Hydrogen iodide is extremely soluble in water (distribution coefficient, 9×10^6 at 25°C),[9] and its absorption is gas-film controlled.

2. On the other hand, methyl iodide is slightly soluble in water (distribution coefficient ranges from 1 to 4).[10] Because of this, we find that its rate of absorption is controlled by liquid film resistance and/or by the kinetics of liquid-phase reactions.

3. Removal of particles by sprays is a much more complex process than removal of chemical compounds. We are no longer dealing with a substance with a single well-defined set of physical and chemical properties, but with a range of sizes and perhaps compositions. Four or five different mechanisms are involved in the removal of particles by sprays.[11,12] One mechanism is interception of particles by the spray drops; this has an efficiency of approximately zero for particles of the sizes likely to be of concern. A second is Brownian diffusion of the particles; diffusion coefficients generally are too low for this to make a significant contribution to the removal rate. A third is diffusiophoresis (motion of the particles to the drop surface along with, for instance, water vapor being condensed on the drop). This may be significant. A fourth is thermophoresis (motion of the particles toward the colder surface in a temperature gradient). This will be significant whenever a temperature gradient exists. Finally, the particles may function as condensation nuclei, and water vapor droplets may form on them. Drops so formed grow large enough to be removed at a significant rate by impaction or by settling out. This is likely to be the most important mechanism of all. In general, our examination of the literature and our experience lead us to believe that sprays will probably remove particles from air-steam atmospheres rapidly enough to be of interest. On the other hand, it probably will be extremely difficult to develop an analytical model describing particle removal.

A good deal of data accumulated over the years on the release and transport of iodine from overheated or melted fuels shows that iodine vapor is the predominant form released,[13–25] even

---

[7] T. K. Sherwood and R. L. Pigford, *Absorption and Extraction*, 2d ed., McGraw-Hill, New York, 1952.

[8] G. Astarita, *Mass Transfer with Chemical Reaction*, Elsevier, New York, 1967.

[9] R. H. Perry, C. H. Chilton, and S. D. Kirkpatrick (eds.), *Chemical Engineers' Handbook*, 4th ed., McGraw-Hill, New York, 1963.

[10] L. C. Schwendiman, J. Mishima, A. K. Postma, L. L. Burger, and R. A. Hasty, *The Washout of Methyl Iodide by Hydrazine Sprays*, BNWL-530 (December 1967).

[11] C. N. Davies (ed.), *Aerosol Science*, Academic, New York, 1966.

[12] N. A. Fuchs, *The Mechanics of Aerosols*, rev. ed., Macmillan, New York, 1964.

[13] T. H. Row and J. L. Wantland, *The Behavior of Iodine in Air Atmospheres – a Résumé of the First Seven Runs Conducted in the Nuclear Safety Pilot Plant*, ORNL-4050 (April 1967).

[14] L. F. Parsly and T. H. Row, *Study of Fission Products Released from Trace-Irradiated UO_2 into Steam-Air Atmospheres (Nuclear Safety Pilot Plant Runs 8 and 9)*, ORNL-TM-1588 (May 1966).

[15] L. F. Parsly and T. H. Row, *Behavior of Fission Products Released from Synthetic High-Burnup UO_2 in Steam Atmospheres (Nuclear Safety Pilot Plant Runs 10–12)*, ORNL-TM-1698 (February 1967).

[16] L. F. Parsly and T. H. Row, *Behavior of Fission Products Released into a Steam-Air Atmosphere from Overheated UO_2 Previously Irradiated to 20 000 Mwd/T (Nuclear Safety Pilot Plant Run 14, Part I)*, ORNL-TM-1908 (September 1967).

[17] *Nucl. Safety Program Semiann. Progr. Rept. June 30, 1962*, ORNL-3310.

[18] *Nucl. Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401.

[19] *Nucl. Safety Program Semiann. Progr. Rept. June 30, 1963*, ORNL-3483.

[20] *Nucl. Safety Program Semiann. Progr. Rept. Dec. 31, 1963*, ORNL-3547.

[21] *Nucl. Safety Program Semiann. Progr. Rept. June 30, 1964*, ORNL-3691.

[22] *Nucl. Safety Program Semiann. Progr. Rept. Dec. 31, 1964*, ORNL-3776.

[23] *Nucl. Safety Program Semiann. Progr. Rept. June 30, 1965*, ORNL-3843.

[24] *Nucl. Safety Program Semiann. Progr. Rept. Dec. 31, 1965*, ORNL-3915.

[25] G. W. Parker, G. E. Creek, C. J. Barton, W. J. Martin, and R. A. Lorenz, *Out-of-Pile Studies of Fission Product Released from Overheated Reactor Fuels at ORNL 1955–1965*, ORNL-3981 (July 1967).

when significant quantities of particles are present. This was found not only at the Nuclear Safety Pilot Plant[13–16] but also in the in-pile experiments of Miller *et al.*[17–24] and the in-pile and out-of-pile experiments of Parker *et al.*[17–25] In addition, a relatively simple analytical model for the removal of iodine vapor from air by reactive sprays was proposed by Griffiths.[26] We have found that this model is somewhat oversimplified and have extended it to deal with real sprays.[27,28]

Since iodine vapor appeared to be the most important iodine form, since we had considerable experience in handling it, and since we felt we knew which spray solutions would probably be chosen to remove it, we decided to do iodine removal experiments first in the Nuclear Safety Pilot Plant. This report describes experiments which have been done in the NSPP on the cleanup of iodine vapor by sprays and compares the experimental results with theoretical models. The removal of methyl iodide and particles containing iodine will be investigated in future studies.

EXPERIMENTAL EQUIPMENT AND PROCEDURES

The flowsheet of the Nuclear Safety Pilot Plant is given in Fig. 1. The principal component is the model containment vessel (MCV). This is a 10-ft-diam by 15-ft cylindrical vessel with ASME flanged and dished heads designed for 60 psig. Its volume is 1350 ft^3 (38.3 m^3). The furnace is used when UO_2 is to be melted. For the present experiments it was replaced by a steam-jacketed pipe tee for vaporizing iodine.

The decontamination solution storage tank originally was a 55-gal stainless steel drum. However, for our present program we decided we probably would have to run this tank at the same pressure as the MCV and therefore built a replacement vessel and installed it after the first three runs had been made. The new vessel has about twice the capacity of the original drum. It is provided with a large cooling coil so we can remove heat from the solution. For long-duration runs in which the solution must be recycled, we will have to do some cooling to avoid cavitation of the spray pump. The canned motor decontamination solution circulating pump is used to deliver the spray.

At the beginning of the program, a header with 12 Spraco J-140D[29] misting nozzles was installed in the MCV. These nozzles have a capacity of 2.5 gal/hr each at 40 psi differential pressure, and the manufacturer states that they produce 100-μ-diam drops. We used this header during the first three runs in the present series and used a duplex Milton Roy pump to supply the spray header. By that time the new solution storage tank was ready; it was installed, and the original spray header was replaced with a single Spraco No. 1713 ramp bottom nozzle. This nozzle has been proposed for installation in containment buildings being designed. This nozzle is placed on the vessel center line and as high as possible.

---

[26] V. Griffiths, *The Removal of Iodine from the Atmosphere by Sprays*, UKAEA report AHSB (S)R-45.

[27] L. F. Parsly, *Removal of Elemental Iodine from Steam-Air Atmospheres by Reactive Sprays*, ORNL-TM-1911 (October 1967).

[28] L. F. Parsly, *Improved Procedure for Calculating Gas-Film Coefficients for Containment Sprays* (in preparation).

[29] Spray Engineering Co., Burlington, Mass.

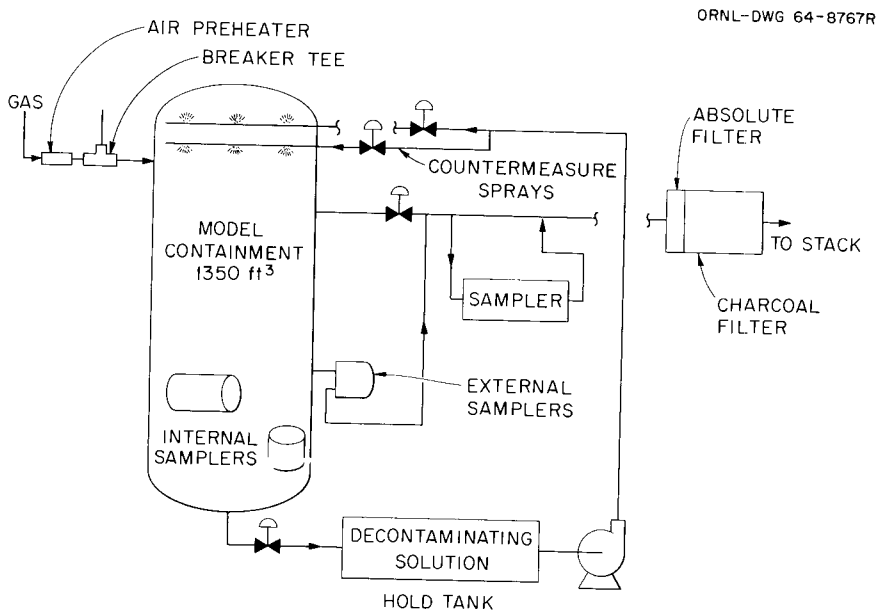

Fig. 1. Nuclear Safety Pilot Plant Flowsheet.

During the first three runs, we used May pack cluster gas samplers[30] placed inside the model containment vessel. Since then, bomb samplers mounted outside the MCV have been used. These are not quite as good as May pack samplers, but using them helps to shorten turnaround time.

At the start of the spray program a 3-gpm canned motor pump was provided to circulate the liquid accumulated in the bottom of the MCV and as a means of getting grab samples of the circulating liquid. This worked fairly well with the misting nozzles and total flows of the order of 0.5 gpm. With the type 1713 nozzle, the flows are too large (9.5 to 15 gpm), for good mixing of the liquid in the bottom of the vessel.

While installing the new storage tank, we also added a sampler to collect liquid falling through the tank atmosphere. It consists of a radial trough extending from 1 in. from the wall to the center and piped to a series of bombs. The sampler was made in this form because calculation of the spray pattern indicated that the point of maximum flow would vary radially with temperature.

We also have a sampler which intercepts liquid running down the tank wall, so that samples of this runoff can be obtained as well as those of solution which fell through the tank atmosphere.

Since preliminary calculations indicated that we might be dealing with half-lives of the order of seconds, we decided to provide a fast gamma-ray recorder as a means of following the transient. Two single-channel analyzers were employed: one of these measured gamma-ray intensity in the vessel; the second measured that in the liquid being circulated to the sampler from the pool

[30] L. F. Parsly, *Experience with May Packs in the Nuclear Safety Pilot Plant*, ORNL-TM-2044 (February 1968).

in the bottom of the MCV. A two-pen Moseley 7100B recorder was used to record the output of these instruments. This instrument permits the chart speed to be varied in steps from 1 in./hr to 2 in./sec. This has proved very convenient; we can record the transient at a relatively high chart speed and switch to a low speed before and after, so that the record is reasonably compact.

The first step in making an experiment is to assemble and load the samplers and make a leak test. We use pressure decay and require that the pressure loss rate be less than 1% in 24 hr with 25 psig in the system.

The iodine is specially prepared by the ORNL Isotopes Division and contains specified amounts of ^{127}I carrier and ^{131}I. The element is prepared by thermal decomposition of PdI_2 in vacuo and is collected in a Pyrex ampoule in a liquid-nitrogen-cooled trap. The ampoule is sealed after it is filled. The ampoule is placed in the steam-jacketed pipe tee using a pair of tongs, and final connections are made. Normally, this step is completed the day before a run, and a final overnight leak check is made with the ampoule in the system.

If a steam-air atmosphere is to be used, the MCV atmosphere is brought to the desired temperature by introducing open steam. The tee is preheated at the same time, and a slight air purge through the tee is used to minimize transfer of steam from the MCV into the tee. If an air atmosphere is to be used in the MCV, it is simply pressurized to 2 or 3 psig with instrument air.

Once the desired pressures and temperatures have been attained, the ampoule is broken by a tool operated through a bellows seal on the branch of the tee. Dry air is used to sweep the iodine vapor into the MCV, and the draft tube in the MCV is operated with steam or air, depending on the atmosphere in the vessel, to mix the iodine. A 10-min period has been used to transfer and mix the iodine.

After the transfer is completed, we take our initial MCV atmosphere sample. After this sample has been taken, we collect initial liquid samples and turn on the spray. The spray flow is adjusted as quickly as possible to the desired rate (usually this can be done in less than 15 sec). During steam atmosphere runs, we have been adding steam while the sprays are operating to minimize the temperature decrease.

Liquid samples are collected at predetermined intervals during the spraying period, and the gamma intensity is recorded. In the early runs, we took May pack gas samples.

In the runs done to date, spraying has ended after 6 min. Generally, there is little change in the gamma intensity measured by the instruments after approximately 2 min. The spray solution may be recycled and spraying continued for long periods of time. We probably will try at least one long-term (24 hr) run to look for possible changes in the solution.

After stopping the sprays, we wait 20 min for the mist to settle and take the final gas sample. The system is then allowed to cool. When the temperature is below 100°C, the solution is drained from the MCV, transferred to a weigh tank, and sampled. After the vessel has cooled below 50° it is purged at a flow of 30 cfm for 4 hr. Our purpose in letting the vessel cool is to avoid venting too much steam to the stack filters. A measured sidestream from the purge is collected on a May pack so that we can determine the amount of iodine purged.

After the system is purged, we enter the hot cell and disconnect and remove the samples.

Liquid samples are bottled, and the sample bombs are rinsed with NaOH solution and water; these rinses also are bottled. Bomb-type gas samples are flushed through a May pack overnight. The purpose is to place any methyl iodide on the second charcoal bed of the May pack (iodine-impregnated charcoal). Then the bomb is decontaminated and rinsed as above.

The MCV is decontaminated by circulating NaOH solution at 60°C over its surface. One decontamination header gives complete coverage of the top head, and a second covers the perimeter of the cylindrical shell at the top tangent. By using these in turn, we wash all but a few spots on the vessel interior. Three water rinses are used. We then decontaminate the tee in place, again using the NaOH and three rinses. Each solution batch is weighed and sampled after use.

The samples are analyzed for iodine by gamma counting. Where useful data can be obtained, conventional chemical analyses are also run for constituents of the spray solutions tested.

Interpretation of Gamma Recorder Data

There is a problem in interpreting the gamma recorder data from the MCV because there is always a significant contribution from iodine plated on the MCV wall in dry runs or in the condensate film in steam runs. In the air atmosphere runs, it has proved fairly easy to establish a background level; the activity present after the spraying period can be taken as background. This does not vary after the spray is turned off. In the steam runs, most of the activity is in the condensate film rather than on the surface and drains off with the condensate. For this case, we decided that the activity at the end of the spraying period was the best measure of the background.

EXPERIMENTAL RESULTS

The experimental conditions for runs 21–22, 26–28, and 30–33 are presented in Table 1. Runs 23 and 29 were not iodine absorption runs, and runs 20, 24, and 25 have been discarded because we had trouble vaporizing the iodine and because too much of it (80% or more) remained in the vaporizer. The table shows that experiments have been done at two temperatures (room temperature and 130°C), using two spray systems, several solution compositions, and two solution temperatures.

Material balances and other experimental results are shown in Table 2. The material balances indicate that we are able to keep a very good account of the iodine present. We believe that the value for the iodine inventory in the ampoule is the least reliable item in the balance, although effort is made to get the best possible measurement of the initial inventory.

Table 1. Iodine Scrubbing Experiments in the NSPP. Operating Conditions

Operating Conditions	Run Number								
	21	22	26	27	28	30	31	32	33
MCV atmosphere									
Temperature, °C	*a*	*a*	130	130	130	130	*a*	130	130
Pressure, psig	3	3	45	45	45	45	3	45	45
Spray solution composition (nominal)									
13.7 g H_3BO_3/liter (2400 ppm B)	x	x	x		x	x	x	x	x
8 g $Na_2S_2O_3$/liter	x	x	x						
3.4 g NaOH/liter		x	x		x	x	x	x	x
pH	7.0	9.2	9.2	7.0	9.2	9.2	9.2	9.2	9.2
Number of nozzles	12	12	1	1	1	1	1	1	1
Type of nozzle[b]	J-140D	J-140D	1713	1713	1713	1713	1713	1713	1713
Flow, gpm	0.57	0.52	10.3	9.9	10.1	15.5	15.3	10.3	10.6
Temperature of spray solution, °C	*a*	*a*	*a*	*a*	120	*a*	*a*	*a*	*a*
Nominal iodine concentration in atmosphere, mg/m^3	5	5	100	100	100	100	100	100	1

[a]Room temperature, approximately 30°C.

[b]Spray Engineering Co. catalog designation.

Table 2. Material Balance

	Run Number								
	21	22	26	27	28	30	31	32	33
Activity balance									
Iodine in source, mc	712	119	239	222	130	225	233	234	246
Iodine in source, mg	800	800	8000	8000	8000	8000	8000	8000	80
Iodine accounted for, mc	649	128	224	269	181	261	224	195	147
Iodine accounted for, %	91	108	94	118	139	117	96	83	60
Distribution of recovered iodine, %									
Spray solution	23.57	40.53	8.79	87.73	88.25	90.23	82.96	88.37	54.01
MCV	34.46	45.35	7.51	10.06	7.03	7.18	10.51	9.05	10.39
Breaker tee	41.83	13.36	10.65	1.52	4.41	2.15	6.48	2.48	33.55
Purge	0.13		0.07	0.37	0.32	0.44	0.04	0.10	2.04

ANALYSES OF DATA

Table 3 presents a summary of our analysis of the experiment. The results are presented in two ways: half-life (time to reduce airborne iodine concentration by a factor of 2) and overall decontamination factor.

We determine the overall decontamination factor by dividing the amount of iodine in the MCV atmosphere at the start of spraying (taken as the sum of the amount picked up by solution and the amount left in the atmosphere) by the amount left in the MCV atmosphere, as indicated by a sample of the air being purged from the vessel. It is necessary to estimate the amount of condensate present just before starting to spray and to calculate the iodine inventory; the amount of condensate is determined as follows:

1. The total spray plus condensate after the run is determined by weighing the condensate.
2. The volume of liquid sprayed is found by gaging the solution storage tank before and after spraying.

The volume present as vapor in the MCV atmosphere can be calculated from the MCV volume and temperature. All liquid in excess of the sum of the volume sprayed and the volume which existed as vapor can be assumed to have been present as liquid when the sprays were turned on.

We have data from two samples to determine the concentration of iodine in the condensate. One is the MCV wall runoff sample; the second is the first of four grab samples of the liquid accumulated in the bottom of the MCV. The latter consistently contains more activity than the former, and we use its concentration as that in the liquid before the sprays are turned on. This choice gives conservative results.

Table 3. Analyses of Data on Iodine Removal by Sprays in the NSPP

	Run Number								
	21	22	26	27	28	30	31	32	33
Overall decontamination factor from solution and purge[a]	169		1858	123	248	243	1800	418	670
Half-lives from experimental data, sec									
Rate of change of gamma-ray intensity	37	38	31	48	48	24	38	41	21
Iodine activity in solution			32	52	45	28	76	46	27, 72[b]
Solution and purge	65		32	50	44	44	32	40	37
Buildup in solution in bottom of vessel	31	35							

[a]To determine the decontamination factor, divide the amount left in the MCV atmosphere (as determined by a cumulative sample of the purge) into the amount left in the MCV atmosphere plus the amount transferred to the solution while spraying.

[b]Two distinct half-lives were observed.

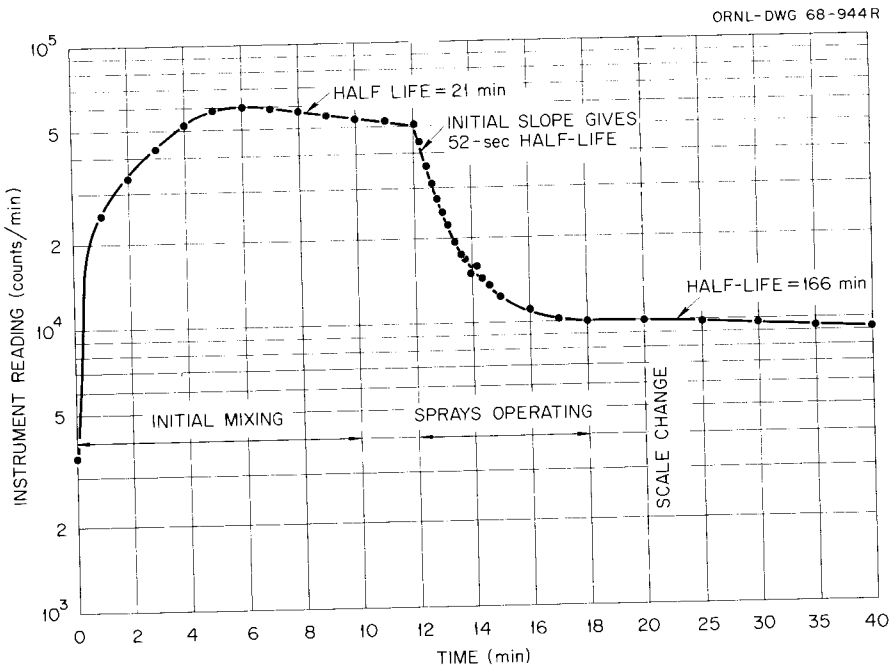

Fig. 2. Uncorrected Gamma Trace from Run 32.

We have applied four different procedures to determine the half-lives. In the first procedure we use the decontamination factor given in Table 3 along with the spraying time to get a half-life value. The second involves the rate of decrease in gamma-ray intensity from the MCV. We have an NaI(Tl) gamma-ray detector mounted near the MCV wall in the upper part. The instrument trace, typically, will be as shown in Fig. 2, which presents the gamma data from run 32. The ampoule containing the iodine was broken at time 0, and the transfer and mixing evidently were completed in 6 min, although air flow from the vaporizer and mixing in the MCV were maintained for up to 10 min. In the interval from 10 to 12 min we took our initial gas sample. The spray was turned on at 12 min and operated for 6 min. The spray was turned off, and after 40 min a second gas sample was taken. The activity level continued to decline during this time, the half-life being about 166 min.

The plot in Fig. 2 shows that the activity at the end of the run was approximately three times that at the beginning. The gamma detector sees activity on the inside wall of the vessel as well as that in the gas, and the wall activity has a relatively high importance because it is so near to the detector. In steam atmosphere runs, such as run 32, this iodine is largely in the condensate film rather than on the vessel surface proper, and the process controlling the reduction in gamma intensity after the sprays were turned off was probably dilution and draining of the condensate film.

We believe that a more realistic value of the half-life for iodine removal by the spray is obtained if we correct the activity level for background. Figure 3 shows data for the spraying period in run 32 using a value of 9800 counts/min for the background. The corrected plot indicates that there was a jump in the gamma reading at 14 min, corresponding to an instrument scale

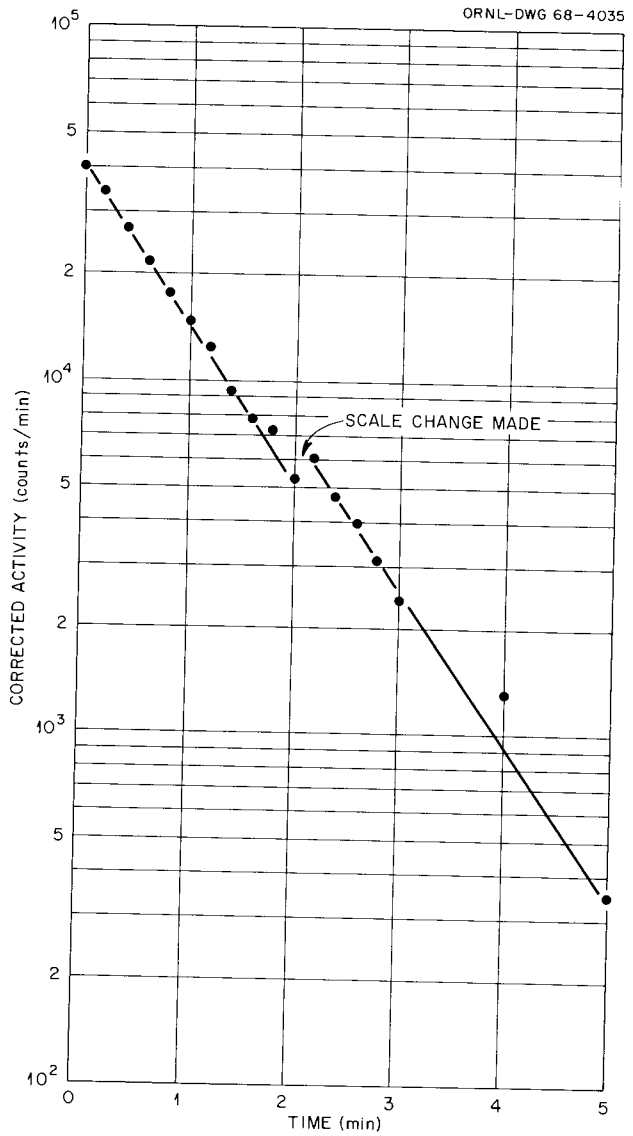

Fig. 3. Gamma Trace for Run 32 with Background Correction.

shift. It gives a half-life of 41 sec. We have used this technique for interpreting the MCV gamma data in all of the runs.

Starting with run 26, there was a series of 1-min samples of the spray solution available. By assuming a first-order removal process and writing a material balance on the containment vessel, we can show that

$$\sum_{i}^{n} \overline{C}_L = B(e^{-\lambda t_i} - e^{-\lambda t_n}) \quad (i = 1, 2, \ldots n) = Be^{-\lambda t_i}, \quad t_i \ll t_n \text{ and } \lambda \text{ large.}$$

Thus a semilogarithmic plot of $\Sigma\, \overline{C}_L$ vs time will have the slope λ, especially for the first few time intervals. This is a third way of determining half-life.

As indicated in Table 2, the results of this procedure generally corroborate those from the MCV gamma-ray intensity.

We also have samples of the liquid running down the vessel wall and of the solution collected in the bottom of the MCV. The former is just condensate in most of the runs, since at the higher temperatures spray reaches the wall only at high flow. The solution collected at the bottom is circulated out of the vessel at a flow of approximately 3 gpm. A gamma instrument was originally provided to measure the intensity in the solution, and we also have four bypass lines which can be provided with solenoid valves at their entrance and exit so that they can be opened and closed remotely and thus can be used to trap samples of the circulating solution. However, we found that at the high spray flows (9 to 15 gpm) we could not mix fast enough to get a representative sample of the solution.

Examination of the tabulated half-life data indicates relatively good agreement among the various methods most of the time. Agreement was poorer in runs 31 and 33. In run 31 a very long half-life was indicated by the gas samples. We see no explanation for this. In run 33, a larger fraction of the iodine quickly deposited in the condensate film, and we probably lost precision due to the relatively high background corrections.

DISCUSSION OF EXPERIMENTS

Effect of Solution Composition

Comparison of the half-life data in Table 2 indicates that there is no obvious effect of the solution on the half-life. This possibly indicates that the partition coefficient for iodine between air and water is significantly higher than the value of approximately 100 which can be inferred from iodine solubility and vapor pressure data. A number of workers[31–33] have produced results which require the partition coefficient to be 10^3 or greater. Eggleton[34] presents a discussion of the iodine-water partition coefficient and of its dependence on concentration which gives some useful clues as to why we might be seeing a large partition coefficient. It would be useful to have partition coefficient data, including the effect of concentration, at elevated temperatures. We are not aware of any such data.

Although the results indicate more favorable performance with water alone than has been expected, we consider it advisable to use an additive which will convert the iodine to a nonvolatile compound.

---

[31] H. R. Diffey, C. H. Rumary, M. J. S. Smith, and R. A. Stinchcombe, "Iodine Clean-Up in a Steam Suppression System," *International Symposium on Fission Product Release and Transport Under Accident Conditions*, CONF-650407, pp. 776–804.

[32] L. C. Watson, A. R. Bancroft, and C. W. Hoelke, *Iodine Containment by Dousing in NPD II*, CRCE-979.

[33] T. Maekawa, Y. Nishizawa, T. Shibayama, and O. Kawaguchi, "Removal of Iodine by Spray Under Atmospheric Pressure," *J. At. Energy Soc. Japan* 7, 563–69 (1965).

[34] A. E. J. Eggleton, *A Theoretical Examination of Iodine-Water Partition Coefficients*, UKAEA report AERE-R-4887.

Effect of Condensation

Condensation on the drops will occur when cold solution is sprayed into a hot saturated atmosphere. One can visualize two possible effects of condensation. If the liquid drops are rigid, a film of condensate can form on the outside; presumably transfer of solute and solution through this layer would be by diffusion. By assuming such a model, one can predict that there would be a significant reduction in the mass transfer rate due to condensation. On the other hand, the flux of vapor to the drop surface can carry along some iodine and increase the mass transfer due to the superposition of the effects of the vapor flux and of diffusion. In five experiments we sprayed hot solution (10°C below the atmosphere temperature) and in one we sprayed cold solution into a cold tank. The data presented in Table 2 show that there are no significant differences in the observed half-life between cases in which there was considerable condensation and those in which there was almost none.

Comparison with Analytical Model

We have been developing an analytical model[28] to describe the performance of sprays. This is an elaboration of the model originally proposed by Griffiths[26] and uses the equation proposed by Ranz and Marshall[35] to calculate gas-film coefficients. Our contributions to the model include the following: (*a*) We calculate physical properties for the atmosphere in which the experiment is being done; (*b*) instead of using a mean drop size, we calculate the performance of a spray which follows a log-normal drop-size distribution; (*c*) instead of assuming that the drops fall at terminal velocity, we calculate an initial velocity based on a considerable simplification of the behavior at the nozzle and from this calculate the trajectory of the drops.

Our model has been programmed in FORTRAN 63 for the CDC 1604 computer. The calculations require approximately 30 sec per case.

Figure 4 presents a comparison of the results of runs 26–33 with the half-lives predicted by our model. This shows that our model predicts half-lives of the right order but does not account properly for the effect of flow. The model indicates that the half-life should decrease with the 2.4 power of the flow, but the data show much less variation.

One reasonable explanation for this which occurs to us is that the drops must be coalescing. The principal reason for the large decrease in half-life with flow is that the mean drop size produced by a nozzle tends to become smaller as flow is increased. Coalescence could result in a different size distribution than expected. To establish whether significant coalescence occurs, it will be necessary to measure drop size as a function of flow at several different distances from a nozzle.

---

[35] W. E. Ranz and W. R. Marshall, Jr., *Chem. Eng. Progr.* **48**, 141, 173 (1952).

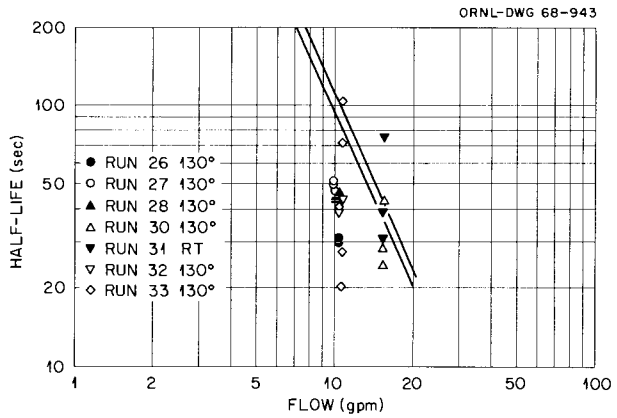

Fig. 4. Comparison of Experimental Results with Analytical Model.

SUMMARY AND CONCLUSIONS

Nine experiments in which iodine vapor with ^{131}I tracer was released into the atmosphere of the model containment vessel of the Nuclear Safety Pilot Plant and then removed by sprays are reported. The half-lives for removing iodine were measured by four independent methods. The half-lives ranged from 20 to 104 sec, with the majority less than 60 sec. Seven of the nine experiments used nozzles and liquid mass flows proposed for use in reactor containment buildings being designed at the present time. Using the same nozzles, the same flow per nozzle, and the same mass flow, one would expect the same performance in a building of any size. Thus we have demonstrated that the half-life for removing iodine vapor from the atmosphere of a containment building can be made 1 min or less (this applies to iodine vapor, *not* to iodine associated with particles or to methyl iodide).

In the series of experiments process water was used in one, borated sodium thiosulfate in four, and borated sodium hydroxide in four. We found little if any significant difference in half-life due to solution composition; we also found that the half-lives were not significantly affected by condensation on the drops.

The results were compared with the predictions of an analytical model we have developed. The model assumes log-normal drop size distribution. It enables calculation of the residence time for each of 61 size groups and calculation of the contribution to an overall removal coefficient for each group. Absorption is considered to be gas-film controlled, and the equation of Ranz and Marshall[35] is used to calculate the film coefficient. Half-lives of the right magnitude can be predicted, although not as precisely as we would like. However, since the model predicts longer half-lives than are observed, we believe it can be used for design.

ORNL-4253
UC-80 – Reactor Technology

INTERNAL DISTRIBUTION

1. Biology Library
2–4. Central Research Library
5–6. ORNL – Y-12 Technical Library
Document Reference Section
7–41. Laboratory Records Department
42. Laboratory Records, ORNL R.C.
43–283. Nuclear Safety Reports
284. R. E. Adams
285. S. E. Beall
286. W. D. Burch
287. W. L. Carter
288. C. W. Collins
289. W. B. Cottrell
290. G. E. Creek
291. S. J. Cromer
292. F. L. Culler
293. F. F. Dyer
294. B. R. Fish
295. M. Fontana
296. A. P. Fraas
297. J. K. Franzreb
298. P. N. Haubenreich
299. P. P. Holz
300. H. B. Hupf
301. P. R. Kasten
302. D. J. Knowles
303. J. O. Kolb
304. C. E. L[illegible]
305. M. [illegible] Lundin
306. R. N. Lyon
307. H. G. MacPherson
308. R. E. MacPherson
309. W. J. Martin
310. H. C. McCurdy
311. J. R. McWherter
312. A. J. Miller
313. P. Newmann
314. E. C. Parrish
315. G. W. Parker
316. R. B. Parker
317–326. L. F. Parsly
327. J. Perona
328. A. M. Perry
329. H. B. Piper
330. B. F. Roberts
331. M. W. Rosenthal
332–371. T. H. Row
372. A. W. Savolainen
373. R. P. Shields
374. M. J. Skinner
375. I. Spiewak
376. L. E. Stanford
377. W. G. Stockdale
378. D. A. Sundberg
379. D. E. Tidwell
380. D. B. Trauger
381. V. R. R. Uppuluri
382. J. L. Wantland
383. G. M. Watson
384. A. M. Weinberg
385. R. C. Weir
386. G. D. Whitman

EXTERNAL DISTRIBUTION

387. L. E. Alsager, DRDT, AEC, Washington
388. E. C. Bailey, Commonwealth Edison Co., Chicago, Ill.
389. R. E. Baker, DRL, AEC, Washington
390. R. E. Ball, Byron Jackson Pump Inc., Box 2017 Terminal Annex, Los Angeles, Calif.
391. G. C. Banick, General Electric Company, Oak Ridge, Tennessee
392. F. Boston, Phillips Petroleum Company
393. H. W. Behrman, AEC Site Representative, Bldg. 4500, ORNL
394. W. G. Belter, DRDT, AEC, Washington
395. D. Black, Idaho Nuclear Corporation, Idaho Falls, Idaho
396. R. A. Birkel, DRL, AEC, Washington

397. R. Boylan, American Air Filter Co., Louisville, Ky.
398. E. B. Branch, Newport News Ship Building & Dry Dock Co., Newport News, Va. 23607
399. Heinz Büchler, Federal Ministry of Scientific Research, Bad Godesberg, Germany
400. J. A. Buckham, Idaho Nuclear Corp., Idaho Falls, Idaho
401. B. Cametti, Westinghouse Electric Corporation, Cheswick, Pa.
402. W. A. Chittenden, Sargent and Lundy, Chicago, Illinois
403. R. L. Cloud, Westinghouse Electric Corporation, Box 79, West Mifflin, Pa.
404. V. Coleman, Pacific Northwest Laboratories, Battelle Memorial Institute, Richland, Washington
405. W. E. Cooper, Teledyne Materials Research Co., 303 Bear Hill Road, Waltham, Mass.
406. J. E. Corr, General Electric Company, San Jose, Calif.
407. H. T. Corten, 321 Talbot Laboratory, University of Illinois, Urbana, Ill.
408. W. D. Doty, U.S. Steel Corp., Monroeville, Pa.
409. F. R. Drahas, Byron Jackson Pump, Inc., Box 2017, Terminal Annex, Los Angeles, Calif.
410. R. J. Edwards, Liquid Metals Engineering Center, Box 309, Canoga Park, Calif.
411. D. A. Eldred, General Electric Company, San Jose, Calif.
412. N. F. Fifer, Douglas United Nuclear, Box 490, Richland, Washington 99352
413. Barbara Frautschi, Information Research Center, Battelle Memorial Institute, 505 King Ave., Columbus, Ohio 43201
414. J. M. Genco, Battelle Memorial Institute, Columbus, Ohio
415. E. F. Gerwin, The M. W. Kellogg Company, Box 881, Williamsport, Pa.
416. J. A. Gieseke, Battelle Memorial Institute, Columbus, Ohio
417. W. D. Gilbert, General Electric Company, San Jose, Calif. 95125
418. J. J. Gough, Code 706, U.S. Navy Marine Engineering Laboratory, Annapolis, Md. 21402
419. J. G. Gratton, DTI, AEC, Washington
420. D. K. Greenwald, Ladish Company, Cudahy, Wis.
421. J. E. Grund, Phillips Petroleum Company
422. L. L. Guzick, U.S. Navy Dept., Bureau of Ships Code 609.36, Standardization Societies, Liaison Section, Washington
423. O. Hagen, Westinghouse Electric Corporation, Cheswick, Pa.
424. R. A. Hasty, Pacific Northwest Laboratory
425. R. L. Heath, Idaho Nuclear Corp., Box 1845, Idaho Falls, Idaho
426. W. G. Hegener, Sargent & Lundy, 140 So. Dearborn St., Chicago, Illinois
427. W. F. Heneghan, Witco Chemical Company, New York, N.Y.
428. R. L. Hervin, ORO, AEC
429. P. G. Holsted, DRDT Senior Site Representative, Box 550, Richland, Wash.
430. N. Horton, General Electric Company, San Jose, Calif. 95125
431. P. W. Howe, DRL, AEC, Washington
432. S. M. Ingeneri, Stone and Webster Engineering Corporation, Boston, Mass.
433. O. H. Jones, Babcock & Wilcox Company, Box 785, Lynchburg, Va.
434. W. G. Knecht, Darling Valve & Mfg. Co., Williamsport, Pa.
435. J. L. Kovach, Barnebey-Cheney, Columbus, Ohio
436. P. Lakey, Idaho Nuclear Corporation, Idaho Falls, Idaho
437. D. F. Landers. Teledyne Materials, 303 Bear Hill Road, Waltham, Mass. 02154
438. B. F. Langer, Westinghouse Electric Corporation, Pittsburgh, Pa.
439. C. F. Larson, Welding Research Council, 345 E. 47th Street, New York, N.Y. 10017
440. A. H. Lazar, Babcock & Wilcox Company, Box 1260, Lynchburg, Va.
441. J. D. Lubahn, Metallurgical Engineering Dept., Colorado School of Mines, Golden, Colorado
442. R. N. Lutman, General Electric Company, San Jose, Calif.
443. M. V. Malkmus, Tube Turns, Div. of Chetrom Corp., Louisville, Ky.

444. R. I. Marble, Farr Company, P.O. Box 90187, Airport Station, Los Angeles, Calif. 90009
445. G. B. Matheny, Phillips Petroleum Company, Idaho Falls, Idaho
446. H. L. McDermot, Defense Research Board, Ottawa, Ontario, Att: Library
447. J. B. McDonough, MSA Research Corp., Evans City, Pa.
448. T. W. McIntosh, DRDT, AEC, Washington, D.C.
449. J. T. McKeon, The M. W. Kellogg Company, Box 696, Piscataway, New Jersey 08834
450. W. N. McLean, Crane Co., 4100 South Kenzie, Chicago, Ill.
451. J. L. Mershon, DRDT, AEC, Washington
452. Walter Mitchell III, Project Manager, Southern Nuclear Engineering, Inc., Box 10, Dunedin, Fla.
453. D. A. Nitti, Babcock & Wilcox Company, Box 1266, Lynchburg, Va.
454. NRTS Technical Library, Box 1845, Idaho Falls, Idaho 83401
455. D. T. Oakley, National Center for Radiological Health, 12720 Twinbrook Parkway, Rockville, Maryland 20852
456. C. S. Patterson, Furman University
457. W. D. Pearl, General Electric Company, San Jose, Calif.
458. D. B. Peterson, Westinghouse Electric Corporation, Box 79, West Mifflin, Pa. 15122
459. A. K. Postma, Pacific Northwest Laboratories, Battelle Memorial Institute, Richland, Wash.
460. N. F. Prescott, Atwood & Morrill Co., Salem, Mass.
461. B. Resnick, Components Branch, DRDT, AEC, Washington
462. R. L. Ritzman, Battelle Memorial Institute, Columbus, Ohio
463. I. C. Roberts, DRDT, AEC, Washington
464. E. C. Rodabaugh, Battelle Memorial Institute
465. G. J. Rogers, Pacific Northwest Laboratory
466. B. J. Round, Combustion Engineering, Windsor, Conn.
467. W. J. Schmidt, Niagara-Mohawk Power Corporation
468. W. Schultheis, General Electric Company, San Jose, Calif.
469. F. R. Schwartz, Jr., North American Carbon, Columbus, Ohio
470. J. T. Sevier, NUS Corporation
471. R. C. Shank, Idaho Nuclear Corp., Box 1845, Idaho Falls, Idaho
472. M. Shaw, DRDT, AEC, Washington
473. H. Sindt, DRDT, AEC, Washington
474. W. L. Slagle, Phillips Petroleum Company, Idaho Falls, Idaho
475. C. M. Slansky, Idaho Nuclear Corporation
476. Craig Smith, Nuclear Energy Laboratory, University of California, Los Angeles, Calif. 90024
477. M. L. Smith, Douglas United Nuclear Co., Richland, Wash.
478. W. R. Smith, Sr., General Electric Company, San Jose, Calif.
479. R. E. Soderberg, Walworth Company, Huff Ave., Greensburg, Pa. 15601
480. J. R. Spink, Douglas United Nuclear, Inc., 1760 D Building, Box 490, Richland, Wash. 99352
481. A. Stathoplos, Nuclear Technology Corp., 116 Main Street, White Plains, N.Y. 10601
482. L. E. Steele, DRDT, AEC, Washington
483. Virginia Sternberg, Westinghouse Electric Corporation, West Mifflin, Pa.
484. J. A. Swartout, Union Carbide Corporation, New York
485. S. W. Tagart, General Electric Company, San Jose, Calif.
486. D. R. Thomas, Commonwealth Associates, 209 East Washington Ave., Jackson, Mich. 49201
487. J. R. Thorpe, First Atomic Ship Transport, Inc., Hoboken, N.J.
488. P. L. Vissat, Taylor Forge & Pipe Works, P.O. Box 485, Chicago, Ill.

489. L. E. Wright, C. F. Brown & Co., Alhambra, Calif.
490. R. R. Wright, Idaho Operations Office
491. J. R. Youngblood, Knolls Atomic Power Laboratory, Schenectady, New York
492. W. A. Yuill, Phillips Petroleum Company
493. Sumio Yukawa, General Electric Company, Schenectady, N.Y.
494. T. Ziebold, Nuclear Engineering Dept., Massachusetts Institute of Technology, Cambridge, Mass.
495. L. Zumwalt, North Carolina State University, P.O. Box 5636, State College Station, Raleigh, N.C. 27607
496. Librarian, Bechtel Corporation, Gaithersburg, Maryland
497. Laboratory and University Division, ORO

498–754. Given distribution as shown in TID-4500 under Reactor Technology category (25 copies – CFSTI)