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CHLORIDE DEPOSITION FROM STEAM ONTO SUPERHEATER FUEL CLAD MATERIALS

Terminal Report
Under Job No. 66-16
Chlorine-36 Deposition Tests

Prepared by

F. Bevilacqua
G. M. Brown

for

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under

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Approved by: _____

J. M. West
J. M. West, Project Director

General Nuclear Engineering Corporation
Dunedin, Florida

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Larry C. Williams
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TABLE OF CONTENTS

I. INTRODUCTION.	1
II. SUMMARY.	3
III. THE Cl^{36} DEPOSITION PROGRAM	10
A. General	10
B. Description of Experimental Equipment and Procedures	10
C. General Description of Test Procedure	13
IV. DESCRIPTIONS OF INDIVIDUAL TESTS	18
A. Tests No. 1 and No. 2	18
B. Test No. 3 - Wet Steam	18
C. Test No. 4 - Repeat of Test No. 3 with Dry Steam	24
D. Chlorine-36 Test No. 5 - Investigation of Temperature Effects	30
E. Chlorine-36 Test No. 6 - Special Surface Roughness Test	37
F. Chlorine-36 Test No. 7 - High Temperature Test	44
G. Chlorine-36 Test No. 8 - Repeat of High- Temperature, Low-Flow Test	51
H. Chlorine-36 Test No. 9 - Special Surface Conditions Test	59
I. Chlorine-36 Test No. 10 - Use of MgCl_2^{36}	72
J. Chlorine-36 Test No. 11 - Successive Additions of Various Solutions of Cl^{36}	79
K. Laboratory Carryover Tests	85
L. Chlorine-36 Deposition Test No. 12 - Use of CuCl_2^{36}	91
M. Chlorine-36 Deposition Test No. 13 - Use of FeCl_3^{36}	95
V. DISCUSSION OF TEST RESULTS	99
VI. CHLORINE-36 INVENTORY RECORD, CONTROL AND DISPOSAL	107

LIST OF TABLES

Table

1	COMPARISON OF TEST PARAMETERS VERSUS BONUS REACTOR PARAMETERS	19
2	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 4	27
3	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 5	34
4	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 6	42
5	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 7	49
6	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 8	55
7	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 9	66
8	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 10	76
9	TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 11	83
10	TABULATION OF DATA FOR LABORATORY BENCH-TYPE CARRYOVER TESTS	87
11	TABULATION OF CARTRIDGE HEATER ACTIVITY LEVELS- TEST NO. 12	93
12	TABULATION OF CARTRIDGE HEATER ACTIVITY LEVELS- TEST NO. 13	97
13	TABULATION OF AMOUNTS OF CHLORINE-36 WHICH DEPOSITED ON THE CARTRIDGE HEATERS	101

LIST OF ILLUSTRATIONS

Figure

- 1 Chloride deposition test equipment layout.
- 2 Sketch of cartridge heater which simulates BONUS superheater fuel rod.
- 3 Detector shield assembly and geometry reference source.
- 4 Over-all view of counting equipment.
- 5 Absorption effect of thin plastic on 0.76-Mev beta particles.
- 6 Absorption effect of thin plastic on 0.71-Mev particles.
- 7 Longitudinal plot of heater activity (Test No. 3).
- 8 Loop effluent (condensate) activity during and after Test No. 4.
- 9 Longitudinal plot of heater activity (Test No. 4).
- 10 Loop effluent concentration during Test No. 5.
- 11 Comparison of cartridge heater activity after Tests No. 4 and No. 5.
- 12 Plot of cartridge heater activity after Test No. 5.
- 13 Photograph of a badly pitted area which contained a heavy deposit of Cl^{36} . (7X)
- 14 Loop effluent concentration during Test No. 6.
- 15 Plot of cartridge heater activity after Test No. 6.
- 16 Loop effluent concentration during Test No. 7.
- 17 Plot of cartridge heater activity after Test No. 7.
- 18 Loop effluent concentration during Test No. 8.
- 19 Plot of cartridge heater activity after Test No. 8.
- 20 Two-plate electrode assembly used during Test No. 9.
- 21 Loop effluent activity during Test No. 9.
- 22 Autoradiograph and photograph of section of heater which experienced wet steam flow.

- 23 Autoradiograph and photograph of intentionally marred heater surface area.
- 24 Autoradiographs of heater surfaces after exposure to steam contaminated with NaCl^{36} .
- 25 Plot of cartridge heater activity after Test No. 9.
- 26 Comparison of same cartridge heater activity levels after two similar tests.
- 27 Loop effluent activity during Test No. 10.
- 28 Plot of cartridge heater activity after Test No. 10.
- 29 Plot of loop effluent activity during Test No. 11.
- 30 Plot of cartridge heater activity after Test No. 11.
- 31 Initial laboratory Cl^{36} release-by-boiling apparatus.
- 32 Laboratory Cl^{36} release-by-boiling apparatus with steam purging provisions.
- 33 Loop effluent activity during Test No. 12.
- 34 Plot of cartridge heater activity after Test No. 12.
- 35 Loop effluent concentration during Test No. 13.
- 36 Plot of cartridge heater activity after Test No. 13.
- 37 Enlarged closeup photograph of typical heater spacer after cleaning.
- 38a Inventory record of Cl^{36} .
- 38b Inventory record of Cl^{36} . (Continued)
- 38c Inventory record of Cl^{36} . (Concluded)
- 39 Diagrammatic description of handling and disposal of Cl^{36} .

I. INTRODUCTION

The BONUS reactor¹ core has an integral superheating fuel zone which uses fuel elements containing UO_2 fuel pellets canned in Inconel tubes; the Inconel clad fuel element is contained in a Type 348 stainless steel coolant tube-pressure tube subassembly which insulates and isolates the fuel element from the surrounding moderator water. At the time that this program was initiated, the UO_2 fuel pellets were also canned in Type 348 stainless steel tubes. Because of various problems (particularly chloride stress corrosion) associated with austenitic steels operating at high temperature and in a steam environment, it became very important to understand the chloride deposition behavior in the BONUS superheaters, in order to determine if catastrophic chloride stress-corrosion failures were likely.

A previous study,² to test the adequacy of the BONUS preheater to prevent chloride deposition and subsequent chloride stress corrosion, had already shown that wet steam containing chlorides and oxygen will cause chloride stress corrosion failure of Type 304 and Type 347 stainless steels. These failures occurred in the tubes of a BONUS preheater mockup being used in the study. This same study also showed that although the BONUS type preheater was effective in removing over 50% of the chlorides, the remaining chlorides would pass through the fuel assembly. Chloride corrosion and deposition data from these tests indicated that no detectable deposition of chlorides had occurred on the test cartridge heaters except in an area where condensation had occurred. This absence of chloride deposition was attributable to the addition of 2 to 6°F of superheat to

the steam which essentially removed all chloride bearing moisture before the steam passed over the test cartridge heaters. However, the tests were of short duration and the chemical analyses for chlorides were probably not sensitive enough to detect a low chloride buildup which may have occurred.

GNEC, therefore, proposed a series of experiments in which radioactive chlorine (Cl^{36}) would be added to the steam used in the test loop. This would allow the amounts of chlorides deposited under various conditions onto superheater clad surfaces to be measured by radioactivity counting techniques. These techniques were much more sensitive than the chemical techniques previously used; deposits containing as little as 1×10^{-8} gms of Cl^{36} could be detected in scale or steam sample residues by using the radioactive tracer technique.

Approval to conduct these experiments as part of the BONUS R&D Program under AEC Contract No. AT(40-1)-2674 was received on May 3, 1962. Test loop operation was initiated in August, 1962 in a test loop situated in, and using saturated and superheated steam obtained from, the Bayboro Plant of the Florida Power Corporation, St. Petersburg, Florida. The tests were completed in July, 1963.

II. SUMMARY AND CONCLUSIONS

In order to determine the deposition behavior of chlorides on the BONUS superheater fuel assembly materials, a series of experiments were conducted in a steam test loop using Cl^{36} as a radioactive tracer contaminant in the steam. The use of Cl^{36} enabled the detection and mapping of as little as 1×10^{-6} gms of Cl^{36} deposited over a 2-ft-long, 3/4-in.-OD cartridge heater and detection of as little as 1×10^{-8} gm of Cl^{36} in heater clad scale or steam sample residue.

All Cl^{36} deposition tests were conducted in an experimental steam test loop installed in Florida Power Corporation's Bayboro Plant, St. Petersburg, Florida. This loop was designed to allow testing of up to four cartridge heaters in steam at a pressure of 800 to 900 psig; steam velocities, cartridge heater clad temperatures, initial steam moisture content or superheat, oxygen content, and chloride content were varied. Generally, three 24-in.-long, 3/4-in.-OD heaters were operated in series during testing. A typical test involved bringing the loop to equilibrium conditions and injecting a solution of NaCl^{36} at 520°F into 875 to 925 psig, 530 to 900°F test loop steam for several hours, and then removing the heaters for examination; occasionally, the free oxygen content of the steam was increased from about 5 ppb to 50,000 ppb.

Each of the tests was conducted by using the following sequence of operations: initially the test loop is brought to the desired equilibrium conditions; a known amount of Cl^{36} salt solution was injected into the steam flow path over a period of several hours of steady-state loop

operation; after injection of the solution was completed, distilled water was added to the solution charging tank and the contents of the tank were injected into the loop to remove any Cl^{36} salt solution remaining in the injection system; test components (cartridge heaters, preheater, electrodes, etc.) were then removed for examination; and the loop was decontaminated by operation with wet steam for several days or weeks. Normally, $3.0 \mu\text{c}$ of Cl^{36} were used for each test.

Several other chlorides besides NaCl^{36} were also tested in the loop to determine their effect on deposition; these were: MgCl^{36} , HCl^{36} , FeCl_3^{36} , and CuCl_2^{36} .

For each of the tests, the incoming steam conditions were adjusted to obtain a given moisture content or a given amount of superheat; the moisture content could be varied between zero and 0.5 wt.% and up to 15°F of superheat could be added to the steam before it passed over the upstream test cartridge heater. Adjustment of the power and steam flow to the cartridge heaters allowed the effect of a broad range of steam and heater temperatures to be studied.

During each test, the desired equilibrium temperature and steam flow conditions were obtained prior to injection of the Cl^{36} solution. The Cl^{36} solution was then injected into the steam flow path over a period of several hours. Samples of the loop effluent were taken at 20 to 30 minute intervals before, during, and after the Cl^{36} injection. Generally, the cartridge heaters were decontaminated by soaking in 1 to 2% HNO_3 and then polishing the heater clad surface with 600-grit emery paper; however, several tests were conducted with different heater clad surface preparations.

The test data on Cl^{36} deposition were obtained by: sampling the loop effluent activity and plotting the activity concentration with respect to time; scanning the test cartridge heaters with a scintillation detector to determine the Cl^{36} deposition pattern; removing and quantitatively analyzing the Cl^{36} collected by each heater; and by examining the Cl^{36} deposition patterns by autoradiography.

The test results indicate that the following typical chloride deposition effects may be expected when 800- to 900-psig steam containing up to 500 ppb of chlorides is passed over heated surfaces:

- 1) Chloride deposition from drying of moist steam results in heavy localized deposits which are adherent. Continued operation with moist steam results in thick layers of chloride deposits. These heavy deposits together with the presence of oxygen and water are conducive to severe chloride stress corrosion of austenitic steels.
- 2) Removal of all moisture from the incoming steam reduces the chloride deposition by a factor of 10 to 20 on newly exposed smooth superheater surfaces; these deposits are of a loose, dust-like nature which are easily removed. Continued operation with dry steam will result in a slow buildup of chloride deposits, the amount of buildup depending mainly on the corrosion of the superheater material surface. Even after long term dry steam operation, previous tests² indicate that the buildup of chlorides will be such that it cannot be detected by chemical analysis. With the complete absence of water and the small chloride buildup, chloride stress corrosion effects are minimized.

- 3) The component surface condition is the second most important variable, and it was found that:
- a) Deposition is definitely increased by surface defects and surface corrosion.
 - b) Surface pits, formed by corrosion or chemical means, appear to have an affinity for chlorides which is several factors greater than the original unpitted surface.
 - c) Simple chemical cleaning procedures are not sufficient for completely removing chlorides from surface pits; this indicates that there is tight adherence of chlorides to surface constituents and that surface reactions may have occurred.
- 4) Steam or heater surface temperatures between 530 and 1100°F, in the range of chloride concentrations used, have little, if any, effect on chloride deposition.
- 5) The degree of steam superheat was not found to have an important effect on chloride deposition, when no moisture existed in the steam. However, the reduction in chloride deposition, gained by the use of slightly superheated steam, may be completely lost if water droplets are carried by the steam or if condensation and water "rundown" occurs in the loop piping.
- 6) Greater deposition occurs at low steam velocities. This appears to be due to a decrease in the mechanical scrubbing action of the steam.
- 7) Spacers and other protuberances generally increase local deposition; this appears to be due to local steam velocity effects, and, in the case of the test specimens, to surface roughness around the protuberances.
- 8) The instantaneous concentration of local steam contamination in equilibrium operation of high-purity systems is materially affected

by loop transients. This is evidenced by the sudden appearance of a fraction of a gram of contaminants, released from component surfaces by temperature or flow variations; this type release represents a very large increase in the normally low equilibrium concentration of contaminants.

- 9) Mechanically-produced surface defects do not appear to collect chlorides as rapidly as corrosion or chemically produced defects.
- 10) The use of different clad materials, such as Inconel, Type 304 and Type 347 stainless steel, did not affect the chloride deposition behavior where the clad materials had similar surface conditions. However, the corrosion effects on the various materials subsequently produce variations in the deposition behavior.
- 11) No definite effect of charged surfaces on chloride deposition was found.
- 12) No significant differences exist between the deposition rates of the chloride salts NaCl , FeCl_3 , or CuCl_2 .
- 13) The deposition of chlorides which are initially in the form of HCl or MgCl_2 is greatly reduced when the steam containing these chloride forms is dried. Drying of either solution produces gaseous HCl .
- 14) Some HCl production should be expected when solutions of soluble chloride salts are heated to dryness; this production is very sensitive to the solution pH and is predominant when low (4.0 or lower) pH solutions are used.
- 15) Autoradiographs can be used to determine the Cl^{36} deposition patterns on test elements.

These results have led to the following conclusions, tabulated below, relative to chloride deposition behavior and the design of nuclear superheater reactor systems:

CONCLUSIONS

- 1) The impingement of moisture on superheater fuel cladding should be prevented. Moisture impingement produces heavy deposits of chlorides which propagate chloride stress corrosion in austenitic stainless steels.
- 2) Small, streamlined spacers should be used for spacing the superheater fuel in the flow path. A definite increase in deposition of chlorides occurs at surface discontinuities such as spacers.
- 3) The use of chemical cleaning or "pickling" solutions on stainless steel surfaces should be avoided. Surfaces produced by chemical cleaning showed a much higher affinity for chlorides than surfaces produced by mechanical polishing.
- 4) Austenitic stainless steel materials which contain surface pits or which are subject to pitting due to chemical cleaning or general corrosion should not be used for superheater cladding. Surface pits tend to accumulate large amounts of chlorides even when the steam is superheated several hundred degrees.
- 5) Transients associated with the operation of systems containing high purity fluids should be minimized. During transients, concentrations of contaminants in the effluents can rise by a factor of 100 or more due to release of the contaminants from component surfaces.
- 6) It is almost impossible to predict the molecular form that chlorides will assume upon the drying of high purity (water) solutions. For instance, an initially low-pH solution will produce some HCl, as will solutions of $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$.
- 7) Steam or clad surface temperatures do not appear to directly affect the deposition of common chloride salts from flowing dry steam onto heated surfaces, where the chloride concentration is in the 0 to 500 ppb range.
- 8) Steam velocities in the 5 to 10 ft/sec range produce more net deposition than velocities in the 70 ft/sec range.
- 9) No feasible procedure exists for the complete elimination of chloride deposition. Even where completely dry steam is used, other effects such as pitting may cause deposition which is

sufficient to produce stress cracking. Certainly this would be expected if moisture were either occasionally or constantly present.

- 10) Additional information is needed concerning the effect of specific compounds of chlorine on stress corrosion and the relationship between drying of steam and the appearance of such compounds.
- 11) While certain procedures and conditions were found to either decrease or increase the collection rate of chlorides by heated components, no means were found which would completely prevent this collection.
- 12) Drying of the steam appears to minimize chloride stress corrosion effects on austenitic stainless ~~steels~~. This conclusion is further evidenced by the satisfactory operation of a Type 348 stainless steel clad BONUS type fuel subassembly in the Vallecitos reactor. This element was exposed to slightly superheated steam from the VBWR for over 80 days at about 13.5 Kw of power, a clad temperature of 1050°F, a heat flux of ~172,000 Btu/(hr)(sq ft), and an exit temperature of 670°F to 680°F; the accumulated exposure was greater than 725 Mwd/ton.

III. THE Cl^{36} DEPOSITION PROGRAM

A. General

The Cl^{36} deposition program was essentially a continuation of the previous study on chloride corrosion effects on BONUS superheater materials.² Because the BONUS superheater fuel clad was initially an austenitic stainless steel, Type 348, (the clad has since been changed to Inconel) some concern existed as to the integrity of this material under long-term operating effects in the reactor. Although the chloride content in the water and steam in the BONUS reactor is to be maintained to a very low value (less than 0.1 ppm) the accumulation of chlorides over a long period of time could lead to chloride stress corrosion.

Data obtained in the previous study² have indicated that drying of the steam removed a considerable amount of the chlorides and that relatively little, if any, of the chlorides were deposited on downstream test heater elements when the steam had been slightly superheated.

Since the chemical techniques which were used in the previous tests were not considered sensitive enough, a test program was initiated using Cl^{36} as a radioactive isotope tracer. The use of Cl^{36} , a beta emitter, allowed the detection of as small as 1×10^{-8} gm of Cl^{36} and therefore the deposition behavior of wet and dry steam containing up to 500 ppb of Cl^{36} on heated surfaces could be easily studied.

B. Description of Experimental Equipment and Procedures

1. Description of Equipment

The Cl^{36} deposition experiments were conducted in a test loop at the Bayboro Plant of the Florida Power Corporation. An isometric view

of the test loop is shown in Fig. 1. The loop is made up of the following details shown on the figure.

- a) The saturated steam supply line (upper left-hand corner).
- b) The centrifugal moisture separator.
- c) The heater used for adjusting the incoming moisture content (H1).
- d) The chloride solution storage tank (lower left-hand corner).
- e) The solution injection pump. This pump is a 0 to 2.0 gph chemical metering pump.
- f) The concentric tube heat exchanger which was used to heat solutions being injected to within a few degrees of the saturation temperature (based on the loop pressure).
- g) The chloride solution spray nozzle. A specially designed and tested nozzle was built in the injection tee to guarantee that thorough mixing of the solution and steam was accomplished at the injection point.
- h) The steam-heated preheater. This unit was installed for a prior test. During these tests, it was used to complete the thermal conditioning of the incoming steam, before the steam was passed over the downstream test cartridge heaters.
- i) The vertical sections of piping which contained the cartridge heaters. There were six positions of this type but, normally, only positions H2, H3, and H4 were used. Position H5 was reserved for the insertion of charged electrodes, corrosion specimens, etc., and H6 and H7 were located in a preheater

bypass loop which was not used during the depositon tests. The arrangement of these test positions allowed downflow and upflow, alternately.

- j) The loop effluent condenser. This unit was used to condense all contaminated steam before it was mixed with the total plant condenser coolant and dumped to the bay. The solid lines in Fig. 1 denote the possibility of contaminated (with Cl^{36}) flow. The dashed lines indicate uncontaminated steam flow. The superheated steam used in the preheater and solution heater was returned uncontaminated to the plant.

Steam condensate samples could be withdrawn from any of the numerous connections S-1, S-2, etc., but generally only the downstream point, S-4, was used. Gaseous oxygen could be metered and added as desired at the chloride injection point.

The cartridge heaters which were inserted into the loop just prior to each test, to simulate BONUS superheater fuel rods, were approximately 30 in. long and 3/4 in. OD. About 24 inches of this length were exposed directly to the steam flow. A sketch of an installed 1/2-in.-diameter heater is shown in Fig. 2; all heaters used in these tests had 3/4-in. diameters. Test heaters were clad in either Inconel, Type 304, or Type 347 stainless steel. They were positioned in the 1-in. Schedule 80 pipe (0.957 in. ID) by three 1/16-in.-diameter longitudinal wire spacers at the bottom or free end, one similar spacer and a 1/16-in.-OD thermocouple assembly near the vertical centerline and near the upper end, and by the supporting flange at the uppermost point.

All piping was generally ASTM A 335, low-alloy material.

B. General Description of Test Procedure

1. Exposure of Components to Contaminated Steam

Each test was generally conducted by: installing the cleaned and polished, or specially prepared cartridge heaters; installing any other components of interest; admitting steam to the loop and adjusting the loop flow, steam temperatures, heater temperatures, etc.; injecting the Cl^{36} solution over a period of several hours; flushing the Cl^{36} injection system with distilled water; and then removing the heaters for examination. During the pre-test loop startup and conditions adjustment, distilled water was added by the Cl^{36} injection pump. This procedure prevented the transients associated with pump startup from occurring during the addition of Cl^{36} .

Before the heaters were re-installed, they were decontaminated by soaking in dilute nitric acid and by mechanical polishing. Occasionally, special heater surface conditions were investigated by exposing heavily oxidized areas, heavily corroded areas, or mechanically or chemically damaged areas to the contaminated steam.

The Cl^{36} was normally injected as a dilute solution of NaCl^{36} . This form was used since the salt water-cooled main turbine condenser is expected to be the principal source of chlorides in the BONUS plant. A solution of either dilute MgCl_2^{36} , HCl^{36} , FeCl_3^{36} , CuCl_2^{36} , or a low-pH solution of NaCl^{36} was injected at least once.*

* The various hydrated forms of the compounds, such as $\text{MgCl}_2^{36} \cdot 6\text{H}_2\text{O}$, are not shown since these data are not available for high-temperature, high-pressure conditions used in the test.

The injection period usually ranged between one and four hours, and about 3.0 μc of Cl^{36} were injected into the loop. Because each microcurie contained about 0.006 gm of chlorine, this addition did not alter the normal plant steam chloride content greatly. For instance, the addition of 1.0 $\mu\text{c/hr}$ to a steam flow of 500 lb/hr only increased the steam chloride content from about 30 ppb to 55 ppb and the chloride content never reached 1.0 ppm. The as-received steam normally contained about 5 ppb of free oxygen; this concentration was occasionally raised to 50,000 ppb by the addition of oxygen from a commercial cylinder. This addition was used in an attempt to obtain a more rapid corrosion rate during the injection period.

Steam condensate samples were taken from the loop effluent before, during, and after each test. Normally, 2000 ml samples were taken during each 20 to 30 minute period. The Cl^{36} in the samples was then precipitated as AgCl^{36} , recovered by filtration, and then counted with a scintillation detector. A detailed description of this procedure is given below in Section III-2.

After the Cl^{36} injection and the injection system flushing process had been completed, the loop was shut down and the cartridge heaters removed for examination; on several occasions the heaters were left in the loop overnight, before removal for examination. This examination included scanning the heater surface at about 50 points for Cl^{36} . A specially arranged scintillation detector was used for this purpose. The examination procedure is covered below in Section III-3. After the examination, the heaters were soaked in glass tubes filled with dilute nitric acid and later polished with emery paper. The Cl^{36} removed during these two steps was then counted.

Test conditions included velocities between 10 and 60 ft/sec, steam temperatures between 530 and 900°F, and heater surface temperatures between 600 and 1200°F. The steam pressure varied with plant load conditions but always remained between 875 and 925 psig.

2. Quantitative Analysis of Steam Condensate and Heater Wash Solution Residue for Cl^{36}

The Cl^{36} was received in 1.0 ± 0.10 μc lots of about 3.0N HCl^{36} . Each of these lots was diluted to 3000 ml in order to obtain a working volume. At this point, one or more 4.0 ml fractions were withdrawn from the 3000 ml working volume, diluted to 2000 ml, and then processed in the same manner as condensate samples taken from the loop effluent stream. This dilution of the 4.0 ml sample to 2000 ml was done in order to obtain concentrations similar to those expected in the condensate samples.

For analysis purposes, 2000 ml samples of steam condensate were normally taken. The pH values of these samples were reduced to 3.0 to 4.0 by the addition of nitric acid, and an excess of silver nitrate was added to precipitate the Cl^{36} as AgCl^{36} ; a very large excess NaCl was added later to produce a relatively large quantity of AgCl to serve as a carrier of the very small amount of AgCl^{36} . This solution was heated to about 190°F. While being held at this temperature, the solution was stirred slowly for 10 minutes. It was allowed to cool before the AgCl^{36} -AgCl was removed by filtering through 11.0-cm-diameter Whatman No. 42 paper; three 1.0-in.-diameter disks were punched from this damp paper for counting. After drying, the three disks were stacked, mounted on a planchet, and their beta activity determined. Because all samples count rates were compared with the count rate of a known quantity of the respective lot of

Cl^{36} , a very accurate analysis of such samples was possible. The heater decontamination solutions were processed in the same manner as condensate samples.

3. Qualitative Examination of Heater Surfaces for Cl^{36}

The cartridge heaters were removed from the steam loop after each test and their surfaces scanned for beta activity. Normally, 48-count readings were taken on each 24-in. active length of a test heater; these readings were spaced at 2-in. intervals longitudinally and at 90° intervals circumferentially. All test data were obtained using the same reference positions to allow direct comparison of all tests.

A special heater and detector holder was fabricated for this purpose. This holder consisted of about three feet of 1-in. Schedule 80 stainless steel pipe and a steel "tee" section shielded with lead. A specific area on the heater jacket was positioned inside the pipe and beneath a window in the lead shielded tee. When the scintillation detector was inserted in the side arm of the tee, its crystal was positioned directly over the window and the heater area to be counted. The window in the pipe was 2.0 in. long and extended 180° around the circumference of the pipe. Photographs of this equipment are shown in Figs. 3 and 4.

A known amount of Cl^{36} was added to a known area of paper on a 3/4-in-OD tube to provide a calibrated, geometrically identical reference source for this work; this source is shown in Fig. 4.

Because the 0.71 Mev beta particles emitted by the Cl^{36} could be absorbed in very thin materials, absorption curves were determined for filter paper and polyethelene plastic in order to quantitatively evaluate such absorption. The absorption plots obtained for Th^{204} (0.74 Mev beta)

and Cl^{36} (0.71 Mev beta) emissions are shown in Figs. 5 and 6, respectively. The absorption coefficients of the standard specimens for the two energies were found to be 0.025 and 0.027 cm^2/mg .

4. Autoradiography Techniques

Autoradiography was used to determine the exact patterns of Cl^{36} deposits which accumulated on several test heater surfaces. To obtain the autoradiograph, the object whose surface was to be examined, such as a cartridge heater sheath, was first covered with a layer of ultra-thin plastic. This plastic was used to protect the unexposed film from mechanical or electrostatic damage. Then a small heater area, normally one to several square inches, was covered with the unexposed film. Two types of film were used, Kodak Ultra-Speed Type-O dental X-ray film and Type KK Industrial X-ray film; the latter is the faster film but is also grainier and is very sensitive to variations in processing procedure.

After positioning the film, the entire area was covered with dark tape and the assembly left in a darkroom for several days or weeks. When the dental film was used, the lead foil screen which accompanies the film was placed on the outer side of the film, beneath the protective tape. After exposure, the film was processed according to the manufacturer's instructions and then examined beneath a microscope or with a slide projector. When desired, the developed film was mounted on finely gridded paper to allow positioning of specific exposed areas of microscopic dimensions.

IV. DESCRIPTIONS OF INDIVIDUAL TESTS

A. Tests No. 1 and No. 2

The first two chloride deposition tests were conducted primarily to perfect the test procedures, check out the loop equipment, and evaluate the problems associated with processing and analyzing condensate samples for Cl^{36} .

Test data, obtained during these tests, indicated that 60 to 75% of the chlorides contained in the steam was initially collected by the mockup of the BONUS reactor preheater. These data confirm previous test data obtained on the chloride removal ability of the BONUS reactor preheater using non-radioactive chlorides.²

B. Test No. 3 - Wet Steam

1. Test Objectives

The objective of this test was to determine the effect of allowing wet steam to impinge on at least one of the three cartridge heaters in the steam test loop. Since each cartridge heater was adding considerable heat to the flowing steam, a spectrum of wet steam, dry (saturated) steam, and slightly superheated steam conditions was experienced by the surfaces of the cartridge heaters.

2. Test Conditions

The test conditions were selected such that at least one or two loop parameters were similar to those of the BONUS reactor. During this test: the steam velocity was slightly lower than the average BONUS superheater inlet velocity but higher than the lowest BONUS velocity; the test

steam pressure was approximately 75 psi lower than the BONUS steam pressure; the test steam contained approximately 0.5% moisture at the inlet to the first heater whereas BONUS steam will be superheated 2 to 5^oF by the pre-heaters; and the cartridge heater had a 0.750-in. OD whereas the BONUS fuel rod has a 0.536-in. OD. A complete tabulation of conditions believed pertinent to chloride deposition is given in Table 1, for the test as well as the BONUS reactor. In general, only BONUS superheater inlet conditions are listed as it is expected that deposition will occur primarily near this point.

Table 1 COMPARISON OF TEST PARAMETERS VERSUS BONUS
REACTOR PARAMETERS

<u>Parameter</u>	<u>BONUS reactor</u>	<u>Test loop</u>
Fuel rod or heater diameter, in.	0.536	0.750
Jacket material	Inconel	Type 347 and Inconel
Length per pass, ft	5.0	2.0
Number of passes	4	3
Coolant channel hydraulic diameter, in.	0.174	0.207
Type of spacer	Bar-on-ring	Brazed-on-wire
Surface condition	Smooth, highly oxidized	Comparatively rough, oxidized
Average steam inlet velocity, ft/sec	65	59
Maximum steam inlet velocity, ft/sec	80	59
Minimum steam inlet velocity, ft/sec	53	59
Heat flux range, Btu/(hr)(sq ft)	55,000 to 201,000	35,000
Pressure, psia	960	875 to 900
Steam moisture content, wt. %	Superheated 2 to 5 ^o F	0.4 to 0.5
Steam temperature, ^o F	543 to 546	529 to 531

3. Test Procedure

The general test procedure followed that given in Section III-B. Initially, wet steam was passed through the loop at the desired rate, the preheater was placed into operation (by admitting 850°F steam), one steam conditioning heater and three test cartridge heaters were turned on, and all loop conditions were adjusted to obtain the values listed in Table 1.

During environmental condition stabilization, the chloride solution charging pump was placed into operation but only distilled water was injected into the loop. This water was heated to between 515 and 525°F before being sprayed into the test loop steam at a point upstream from the preheater; the steam saturation temperature at this point was 531°F. Several feet of piping and one 90° turn were traversed by the water-steam mixture before being passed over the preheater.

During this test a combination of preheater tube-side flow (superheated steam) and a cartridge type steam conditioning heater was used to obtain 0.4 to 0.5% moisture at the inlet to the first test cartridge heater. This moisture content was determined by increasing the power to the steam conditioning heater and noting when a temperature increase occurred at a downstream thermocouple. Since the steam contained some moisture as it left the preheater, wet conditions existed in this area of the loop at all times during this test.

After equilibrium conditions had been attained throughout the loop, one microcurie of NaCl^{36} was diluted to 6.0 liters and added to the solution charging tank. This solution was then injected into the flowing steam over a period of approximately 90 minutes. After this solution had passed

through the injection system, distilled water was added to the charging tank and then to the loop for another 90-minute cleanup period. Following the cleanup period, the charging pump was stopped, the loop steam flow was reduced from about 805 lb/hr to about 150 lb/hr, and the preheater tube-side steam flow was increased. This operation was desired to maintain low velocity, dry steam flow within the loop throughout the night. One microcurie was added each day until a total of three microcuries had been injected.

Samples of steam condensate were collected at the loop exit before testing commenced, during all distilled water injections, during all Cl^{36} injections, after the loop had been secured for the night, and before the loop was disturbed the next day. This procedure was designed to determine the amount of tracer material which flowed directly through the loop and the rate at which it left the loop between injections, in order to obtain a complete Cl^{36} inventory and dumping record.

After injection of the third microcurie of Cl^{36} and the 90-minute cleanup period, using distilled water, the preheater and three cartridge heaters were removed for counting; the empty loop was placed into operation with wet steam being passed through the loop as part of a decontamination process.

During pre-test, test, and post-test operation, all loop conditions such as flow, heater power, and preheater tube-side flow were held constant. The only condition which changed slightly was the steam supply (plant) pressure. This pressure may have varied as much as ± 10 psi during the test. However, this relatively minor variation was not believed to be significant since it did not cause noticeable flow or temperature variations.

4. Test Results

The test data were obtained by taking condensate samples at the loop exit before, during and after the test; by scanning the three cartridge heaters after the test; and by counting all removable Cl^{36} on the three cartridge heaters and preheater. In addition, condensate samples were taken while wet steam was being passed through the loop after the preheater and cartridge heaters had been removed.

It was found that only about 25% of the injected Cl^{36} ($3.0 \mu\text{c}$) passed directly through the loop. The remainder adhered to preheater and other component surfaces, with about $0.55 \mu\text{c}$ leaving the loop between injections, under dry steam conditions, and about $0.007 \mu\text{c}$ leaving under wet steam or decontamination conditions. The majority of the remaining activity was found on the preheater.

The most important and interesting data were obtained by scanning the three cartridge heaters. The upstream heater, which experienced wet flow over its upper (inlet) end, showed high Cl^{36} activity up to a point 6 in. below the steam inlet and exhibited considerably less activity over the remainder of its length. It is significant to find that the major activity occurred within the initial 7 to 8 inches which is the calculated heater distance wherein bulk steam conditions should have progressed to saturation conditions or dryness. A plot of the average circumferential activity along the length of the test heaters is shown in Fig. 7. It can be seen from these data that the major activity existed on areas subjected to wet steam flow and on areas about the spacers. Washing and cleaning in a 2% nitric acid solution removed $0.014 \mu\text{c}$ from the first heater.

No activity was found on the second heater during the scanning procedure; however, the chemical cleaning process removed $0.0006 \mu\text{c}$ from the cladding. It should be noted that this second heater was exposed only to upflow, dry, slightly superheated steam; the inlet steam to the heater was superheated about 12°F and the outlet steam from the heater was superheated about 34°F .

A small amount of activity (see Fig. 7) was found on the third heater, primarily around the thermocouple connection which was positioned about 12 in. from the bottom end of the heater. However, since this heater experienced downflow and upon visual inspection it was evident that some condensation and run-down had occurred on the upper non-heated section, the activity detected on this heater was partially attributed to condensation rather than simple deposition; condensation was attributed to poor insulation of the cartridge heater flange which allowed cooling of the upper end of the heater. Washing and acid solution cleaning removed $0.0014 \mu\text{c}$ from this heater.

The preheater tube bundle (nine smooth U-tubes) was removed and cleaned immediately after the Cl^{36} injection. Approximately $0.97 \mu\text{c}$ of Cl^{36} were removed at this time. In addition, approximately $0.010 \mu\text{c}$ were removed from the preheater shell by scraping and brushing in place; this scraping process was only about 50% effective in removing all of the activity.

The procedure discussed above accounted for approximately 2.9 of the $3.0 \pm 0.3 \mu\text{c}$ of Cl^{36} used in the test. During the testing, almost $2 \mu\text{c}$ of NaCl^{36} were passed over the three cartridge heaters. Of this

amount, the first heater collected about 0.5%, the second heater collected about 0.02%, and the third collected about 0.05%. While the first heater collected 10 to 20 times as much as either of the two remaining heaters, it should be remembered that this ratio would probably have been greater if the operation under wet, chloride containing steam conditions, had been continuous instead of being interrupted by dry "clean" steam operation for two nights. Any operation under dry, "clean" steam conditions is believed to remove a fraction of deposited chlorides.

5. Discussion of Test Data

The following conclusions were drawn from the results obtained in Test No. 3.

- a) Drying of wet steam results in relatively heavy deposits of chlorides.
- b) The chlorides have an affinity for areas about spacers and other protrusions.
- c) A very small fraction of the chlorides adheres to almost all surfaces, even under dry, relatively high velocity conditions.
- d) The chloride deposits tend to be removed by the passage, over the deposits, of wet uncontaminated steam.

C. Test No. 4 - Repeat of Test No. 3 with Dry Steam

1. Test Objectives

The purpose of this test was to obtain test data on chloride deposition behavior under slightly superheated steam conditions, for comparison with the chloride deposition behavior under the wet steam condition of Test No. 3. Similar steam velocities, heater power levels, etc., were used during this test as had been used in Test No. 3.

2. Test Conditions

As mentioned above, the conditions for this test closely duplicated these of Test No. 3 except for inlet steam moisture content. During Test No. 4, the steam was superheated 4 to 6°F before being passed over the first test heater; a combination of preheater tube-side steam flow and conditioning-heater power was used to obtain this condition. Because of the slightly higher steam inlet temperature, coupled with the absence of moisture, the steam and heater surface temperatures were approximately 10 to 15°F higher than the respective temperatures obtained in Test No. 3.

3. Test Procedure

The test procedure for this test was similar to that of Test No. 3 except that all 3 µc of Cl^{36} were injected within one day. The procedure again consisted of: a pre-test stabilization period of environmental conditions; injection of the solution of NaCl^{36} over a period of several hours; and a post-test injection system cleaning operation. The first and last steps involved loop operation with the injection system adding distilled water (instead of NaCl^{36} solution) to the loop.

After the injection system cleaning process was completed (approximately 2 hours operation), the loop was shut down and the cartridge heaters removed for monitoring and cleaning. However, the preheater was left in the loop for about 60 hours of post-test operation before being removed and counted; test conditions were maintained during this operation.

The three pertinent operational differences between Test No. 3 and No. 4 are summarized below.

- a) In Test No. 3 wet steam impinged upon the upstream heater.

In test No. 4 dry steam impinged upon the upstream heater.

- b) In Test No. 3, the loop was placed under dry steam, low velocity conditions overnight (2 nights) and one microcurie of NaCl^{36} was injected each day for three days. In Test No. 4, the 3 μc of NaCl^{36} were injected in one day.
- c) In Test No. 4, the preheater was allowed to remain in the loop for approximately 60 hours at test steam conditions following the cleaning period. In Test No. 3, the preheater was removed immediately after the cleaning period which followed the Cl^{36} injection.

4. Test Results

Test data were obtained by taking condensate samples at the loop exit before, during, and after the test; by scanning the three heaters after the test; and by counting all removable Cl^{36} on the three heaters and on the preheater.

It was found that about 30% of the 3.0 μc of Cl^{36} injected passed directly through the loop. The remainder adhered to the preheater and other component surfaces, with about 2.0 μc leaving the loop after completion of the test and a small amount leaving under wet steam or cleaning conditions. The major portion of these data are shown in Fig. 8.

Once again, the most important and interesting data were obtained by scanning the three cartridge heaters. The upstream heater, which experienced dry, slightly superheated steam flow over its entire surface, exhibited a small amount of activity over its entire length. A tabulation of this activity is shown in Table 2 and a plot of the average circumferential

Table 2 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 4*

Angle from reference, °	Distance from inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
<u>Upstream or first heater (downflow)</u>													
0	140	135	153	168	143	158	155	117	146	118	124	132	133
90	135	189	152	164	138	133	146	137	123	150	129	148	122
180	126	155	168	141	158	122	149	118	116	120	116	132	121
270	109	145	160	157	139	117	144	124	128	104	108	138	143
<u>Second heater (upflow)</u>													
0	-	135	110	131	133	110	128	132	130	192	162	149	131
90	-	113	111	133	130	144	138	146	161	122	142	135	147
180	-	138	122	135	155	139	140	166	127	121	126	125	116
270	-	138	139	111	143	125	132	143	146	157	153	140	144
<u>Downstream or third heater (downflow)</u>													
0	98	136	119	99	127	140	172	196	254	251	304	372	298
90	83	119	127	139	164	161	158	170	190	219	238	299	218
180	118	113	162	159	173	185	147	198	176	181	209	273	246
270	112	117	134	132	161	138	245	197	199	257	288	383	414

* Includes 80-105 counts for background.

activity found along the length of each heater is shown in Fig. 9. From this figure, it can be seen that the activity deposition on the first heater occurred in a somewhat uniform pattern except for discontinuities due to spacers and thermocouples. Soaking in a cold, distilled water bath removed $0.00068 \mu\text{c}$ from this first heater. A later 2% nitric acid wash with scrubbing removed another $0.00026 \mu\text{c}$. It is interesting to note that almost 72% of the active deposit existed as either a loose solid or was soluble in the cold water.

The amount of activity found on the second heater during the scanning procedure was similar to that found on the first heater. The acid cleaning process removed $0.00056 \mu\text{c}$ from this heater which had also experienced dry flow; no cold water wash was used. The degrees of superheat at the heater inlet and outlet were approximately 26 and 48°F , respectively.

Surprisingly, the greatest amount of activity was found on the last (most downstream) heater (see Fig. 9). The activity measured was very low near the inlet end but increased somewhat linearly to a very high value near the outlet end. This heater, like the first heater, was soaked overnight in a distilled water bath, and then overnight in a 2% nitric acid solution. The distilled water removed $0.00311 \mu\text{c}$ while the acid solution removed only $0.000332 \mu\text{c}$. Once again, the removal of a major portion of the activity by soaking in distilled water indicated that the Cl^{36} was either deposited in a loose, dust-like form, or was reasonably soluble in room-temperature water.

The preheater tube bundle (nine smooth U-tubes) was removed about 60 hours after completion of the Cl^{36} injection and was cleaned. Approximately $0.000126 \mu\text{c}$ of Cl^{36} was removed from the tube bundle by soaking in distilled water and scrubbing. No detectable activity was removed by scraping and brushing the preheater shell in place.

The procedure discussed above accounted for approximately 2.95 of the $3.0 \pm 0.3 \mu\text{c}$ used in the test. During the test, about $2 \mu\text{c}$ of NaCl^{36} had been passed over the three heaters. Of this amount, the first heater collected about 0.047%, the second heater collected about 0.03%, and the third heater collected about 0.17%. A plot of loop (condensate) effluent activity data taken during and immediately after the injection is shown in Fig. 8. Data were obtained for an additional two days beyond the time shown on Fig. 8; at the end of the three days an activity level was not detectable.

5. Discussion of Test Results

The following general results were noted after the data for Test No. 4 were reviewed.

- a) The Cl^{36} deposition on the upstream heater was considerably less than that found to occur during Test No. 3. The activity deposited was reduced from $0.014 \mu\text{c}$ (Test No. 3) to $0.00094 \mu\text{c}$ (Test No. 4); this reduction is attributed to the dry or slightly superheated steam used in Test No. 4. It should be noted that the absolute value of the Cl^{36} deposited on the upstream heater during Test No. 3 may have been even greater than the amount removed, due to the intermittent test procedure used.

- b) The continued affinity of chlorides for areas about spacers and thermocouples was again noted. While the scanning equipment being used is not directional to the point that these deposits can be outlined in detail, it is clearly evident that such attachments do cause increased local deposition.
- c) Some chlorides are deposited, at least temporarily on all heated surfaces. While data obtained using conventional analytical procedures were indicative of some deposition, the measuring techniques were not accurate enough to yield conclusive data.² However, the Cl^{36} tracer technique revealed that some chloride deposition occurs on all surfaces and that under dry steam conditions the deposits which occur are in a very loosely-held form.
- d) A pronounced buildup on the third and hottest heater occurred during the test. Initially, the heavy buildup of Cl^{36} on the third or downstream heater was believed to be due to a temperature effect and a test (Test No. 5) was run to investigate the effect of temperature on deposition. However, it was later found that the activity (and deposit) was proportional to the number of pits in the specific areas of this heater.

D. Chlorine-36 Test No. 5 - Investigation of Temperature Effects

1. Test Objectives

This test was designed to investigate the possibility of a temperature effect on Cl^{36} deposition as indicated by the chloride activity measured on the third heater after the completion of Test No. 4.

2. Test Conditions

The heater power levels, pressures, etc. used in Test No. 4 were duplicated except that the steam flow rate was reduced from 810 lb/hr to 605 lb/hr, thus reducing the velocity about the heaters from 61 ft/sec to about 47 ft/sec and increasing the heater and steam temperatures approximately 20°F higher than those used in Test No. 4. This procedure produced temperatures on the second heater which were comparable to those on the third heater during Test No. 4. The preheater was used to dry and add 4 to 8°F of superheat to the incoming steam. The three cartridge heaters then added approximately 85 additional degrees to the steam temperature. Steam entered the first heater section of the loop at 535 to 539°F and exited from the third heater at 620 to 630°F; heater surfaces varied between 560 and 700°F. No wet steam was allowed to impinge on the heaters at any time during this test.

3. Test Procedure

In general, the test procedure duplicated that for Test No. 4. A pre-test stabilization period was run, the NaCl³⁶ was injected over a period of several hours, and then a post-test injection system cleaning operation completed the test. The first and last steps involved loop operation with the injection pump adding distilled water to the steam flow. After the 2-hour cleaning operation was completed, the loop was shut down and the three cartridge heaters were removed for inspection, counting and cleaning. During Test No. 4, the preheater was left in for an additional 60 hours of operation (without Cl³⁶ addition) before being removed. For this test, Test No. 5, the loop was shut down overnight and

the preheater removed the following day. As in Test No. 4, all 3 μc of Cl^{36} were added during one day.

4. Test Results

The test data were obtained: by collecting condensate samples at the loop exit before, during, and after the NaCl^{36} addition; by scanning the heater surfaces after removal from the loop; and by counting all Cl^{36} removable from the heaters and preheater. As before, steam condensate samples were collected for several days.

It was found the approximately 30% of the 3.0 μc passed through the loop during the injection. A plot of the effluent concentration is shown in Fig. 10. This plot is similar to the one obtained for Test No. 4 except that:

- a) The plot of Test No. 5 indicated a disturbance during the first two hours of the injection. This is believed to have been due to a large plant load decrease which resulted in a pressure drop from 920 to less than 900 psia.
- b) The amount of activity which left the loop following loop restart after Test No. 5 was completed (and the three heaters and preheater were removed) is less than the amount of activity which left the loop following loop restart after Test No. 4 was completed (and the three heaters only were removed). This indicates that operation with uncontaminated steam during Test No. 4 washed some of the Cl^{36} deposits from the preheater.
- c) When compared to the data from Test No. 4, data from Test No. 5 showed a higher initial concentration of Cl^{36} in the condensate

samples resulted during re-starting of the loop after the removal of the preheater and heaters. This was apparently due to the longer shutdown and resultant increased flow of wet steam upon re-starting. The uncontaminated wet steam tends to remove a larger fraction of deposits than does dry steam.

Scanning the three cartridge heaters with the scintillation detector provided excellent comparative data. It was found that the deposition pattern shown on the downstream or third heater after Test No. 4 had been repeated very closely for the same physical heater during Test No. 5 even though (1) the steam and surface clad temperatures had been increased and (2) the downstream or third heater from Test No. 4 had been moved to the second heater position where it was subjected to upflow instead of the downflow of Test No. 4. The activity found along the length of the heater after Tests No. 4 and No. 5 is shown on the curves of Fig. 11.

While Test No. 5 produced a slightly heavier deposit, the similarity between the two plots indicated that a contributing factor was involved other than steam conditions or heater temperatures. The heater surface was then examined carefully with a microscope. This examination revealed that the heater surface was pitted severely, with the pits apparently becoming deeper and more numerous as the examination progressed from the top of the heater to the bottom end. A tabulation of the Test No. 5 scanning data for all three heaters is given in Table 3.

As a means of checking the looseness of the deposit on the heavily contaminated heater, various methods of removing the deposit were used.

Table 3 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 5*

Angle from reference, °	Distance from inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
<u>Upstream or first heater (downflow)</u>													
0	109	175	155	155	159	152	142	121	110	126	138	128	170
90	105	129	155	148	123	135	131	126	113	132	121	133	129
180	127	124	115	118	111	119	151	123	109	113	100	115	120
270	129	125	149	150	130	128	158	113	93	116	116	105	146
<u>Second heater (upflow)</u>													
0	-	191	315	306	261	279	247	258	167	178	117	160	122
90	-	180	254	251	273	280	294**	221	206	200	194	145	121
180	-	174	355***	277	243	264	253	277	262	237	262	156	106
270	-	218	312	283	287	240	266	311	233	199	196	156	82
<u>Downstream or third heater (downflow)</u>													
0	-	97	106	110	127	112	118	104	111	115	101	116	130
90	-	93	128	99	114	113	120	117	110	131	126	120	134
180	-	118	101	107	112	108	108	107	118	121	105	119	115
270	-	112	114	125	121	107	121	93	97	122	97	110	108

* Background = 93-105 counts.

** Reduced to 190 after water rinse and 1% HNO₃ soak, 145 after brush scrub.

*** Reduced to 161 after water rinse and 1% HNO₃ soak, 147 after brush scrub.

The results were as follows: a 5-minute dip in cold water removed $136 \times 10^{-4} \mu\text{c}$; a scrubbing wash in a 1% HNO_3 solution removed an additional $4.7 \times 10^{-4} \mu\text{c}$; and a 24-hour soaking and scrubbing in 1% HNO_3 removed an additional $11.6 \times 10^{-4} \mu\text{c}$. The total amount removed from the heater was $152 \times 10^{-4} \mu\text{c}$, as compared to a total of $35 \times 10^{-4} \mu\text{c}$ removed after Test No. 4.

The same three-step cleaning process was used on each of the other two test cartridge heaters. These steps removed 3.4×10^{-4} , 15.3×10^{-4} , and $0.9 \times 10^{-4} \mu\text{c}$ respectively, for a total removal of $19.6 \times 10^{-4} \mu\text{c}$ of Cl^{36} from the upstream test heater; and 1.5×10^{-4} , 3.7×10^{-4} , $1.5 \times 10^{-4} \mu\text{c}$ respectively, for a total removal of $6.7 \times 10^{-4} \mu\text{c}$ of Cl^{36} from the downstream test heater. For each of these two heaters, the water dip removed an intermediate amount of the chloride, the acid wash and scrub a larger amount, and the acid soak an intermediate amount. It should be noted that, for the pitted heater which was positioned between the other two heaters, the greatest amount of chlorides was removed in the water dip. This was apparently due to the loose adherence of the larger amount of chlorides in the severe pits of the test heater.

A plot of the activity found on all three heaters is shown in Fig. 12. This plot is similar to the one obtained for Test No. 4 (Fig. 9) except that the downstream heater in Test No. 4 was moved to the mid-position in the loop during Test No. 5. It is evident from these data that the downstream heater, which was new at this time, collected the least amount of chlorides.

Washing and scrubbing in a 1% HNO_3 solution removed about 0.38 μc from the preheater tubes. These data, coupled with Test No. 4 data where no activity was found 60 hours after test completion (when the preheater was actually removed), indicate that the amount of NaCl which the preheater collects and/or then releases is a function of operation. Apparently, where this soluble compound (NaCl) is deposited on surfaces without being diluted by other scale-forming compounds, it can be generally removed by washing with high-temperature, wet steam.

Although the sampling procedure described above accounted for only 2.7 of the three microcuries which were injected, this is believed to be within the limits of measurement accuracy for the small concentration involved.

Of the 3 μc added, approximately 1.3 μc passed over the cartridge heaters in the loop before the cartridge heaters were removed. Of the 1.3 μc , the upstream heater collected approximately 0.15%, the next heater collected 1.17%, and the downstream heater collected 0.052%. These differences are believed to have resulted primarily from surface conditions.

5. Discussion of Test Results

The following conclusions were drawn concerning the data taken for Test No. 5.

- a) The results of this test indicated that the high concentration seen on the downstream heater after Test No. 4 was due to conditions other than surface temperatures. Severe pitting of the test heater surface was due to earlier high-temperature corrosion work (1000-1200°F) which apparently produced a pitting condition whose severity increased as the test

temperature increased along the heater.

- b) The tests continued to indicate a noticeable affinity of chlorides for areas about spacers, thermocouples, etc.
- c) The effect of surface roughness, especially where pitting is concerned, was obvious. The most heavily pitted areas could be easily located by the activity deposited as indicated in Table 3. A photograph of one of the worst pitted areas on a test heater is shown in Fig. 13.
- d) A major portion of the chlorides deposited under dry steam conditions is in a loose form. This is evident by the large fraction removed by a quick dip of the heaters in cold water. However, a small fraction had apparently reacted with the metal heater surfaces and could not be completely removed even by soaking in nitric acid.

E. Chlorine-36 Test No. 6 - Special Surface Roughness Test

1. Test Objectives

This test was designed to investigate the effect of heater surface roughness on chloride deposition. This was accomplished by repeating Test No. 4 after several new finishes had been applied to the badly pitted heater and a new-tube finish applied to the remaining two heaters.

2. Test Conditions

The test conditions for Test No. 4 were repeated very closely. Steam velocities in the 58 to 62 ft/sec range, steam temperatures in the 535 to 595°F range, and heater surface temperatures in the 550 to 625°F range were used. Once again, the steam was superheated 4 to 8°F by the

preheater before being passed over the cartridge heaters. Pressures ranged from 880 to 920 psig. Several significant plant load changes occurred during the test which resulted in rapidly changing pressures.

3. Test Procedure

The three heaters used during Tests Nos. 4 and 5 were again used during Test No. 6. However, before they were inserted into the loop, the two heaters which were in good condition were buffed circumferentially with 600-grit emery cloth until a mirror-like finish was attained. These two heaters were placed in the upstream (first) and middle (second) positions. The heater with the severely pitted sheath, which had been used in the middle position during Test No. 5, was polished in a special manner and placed in the downstream (third) position, as during Test No. 4.

Several finishes were applied to this third heater. The lower (outlet) 4-in. section was left as before (severely pitted and oxidized); the next 4-in. section was polished with 0.3-micron paste metal polish as well as the 600-grit cloth; the third 4-in. section was buffed with 600-grit cloth; and a fourth section was buffed with 80-grit cloth. It was noted that the pits in this heater were so deep that they could not be removed by this polishing process; they were only reduced in number and size.

The preheater was not removed after the completion of Test No. 6 since it had been previously determined from Test No. 4 that essentially all of the activity on the preheater can be removed by approximately 60 hours of post-test operation.

The procedure used during Test No. 4 was duplicated as closely as possible. A pre-test stabilization period was run, the NaCl^{36} was injected over a period of several hours, and then a post-test injection system cleaning operation completed the test. The first and last steps involved loop operation with the NaCl^{36} solution pump adding only distilled water to the loop steam. As in Test No. 4, after the 2-hour cleaning operation had been completed, the loop was shut down and the three cartridge heaters were removed. Three microcuries of Cl^{36} were used.

4. Test Results

It was found that approximately 37% of the $3.0 \mu\text{c}$ of Cl^{36} passed through the loop during the NaCl^{36} solution injection test period and the subsequent distilled water injection system cleaning period. A plot of the effluent activity is shown in Fig. 14. This plot is similar to those obtained for earlier tests except that the effluent activity level increased slower than during earlier tests and then varied more widely after an apparent equilibrium concentration had been reached. No definite reasons are known for this behavior. However, these effects may have resulted from either steam pressure oscillations or from charging pump output variations; the latter would occur if even minute bubbles or large quantities of absorbed air existed in the NaCl^{36} solution, as the pump only adds a fraction of 1 ml per stroke. In addition, it is clearly evident from Fig. 14 that some Cl^{36} existed in the injection system from previous tests. The exact amount of Cl^{36} remaining from a previous test is a function of the post injection cleaning procedure.

After the cartridge heaters were removed and the loop returned to operation, the injection system was restarted with distilled water in the

tank and loop operation was maintained for approximately 17.5 hours. Thereafter, the loop was left in operation but the injection pump was turned off. The effect of this procedure is also shown in Fig. 14. While the effluent concentration was quite high (about 0.2×10^{-6} $\mu\text{c/ml}$) before stopping the pump, it dropped quite sharply thereafter.

The high effluent activity which persisted as long as the pump was in operation apparently resulted because of release from two sources. One of these was the preheater tube bundle, which was left in the loop after the cartridge heaters had been removed and which had accumulated NaCl^{36} during the test. The other source was the Cl^{36} injection system. The Cl^{36} charging tank had been filled to a relatively high level after the cartridge heaters were removed to allow for overnight operation; this resulted in back-mixing of fluid within the piping with the water in the charging tank, thus producing a large volume of contaminated water of moderately uniform Cl^{36} concentration. In general, when the Cl^{36} injection pump was stopped, the Cl^{36} release from the two sources was generally eliminated. This occurred because the process steam does not pass through the Cl^{36} injection loop piping and because the steam being passed over the preheater was being superheated a few degrees by the upstream conditioning heater and thus did not continue to dissolve the NaCl^{36} deposited on the preheater.

Once again, the most interesting data were obtained by scanning the three cartridge heaters for Cl^{36} activity. The upstream heater, to which a new 600-grit surface had been applied, definitely collected less Cl^{36} than during Test No. 4, i.e., before the polishing. Collection on

the second heater, whose surface preparation was similar to the upstream heater, also showed a decrease in the amount of Cl^{36} collected. However, areas about spacers and thermocouples on both of these heaters exhibited activity levels similar to those found after Test No. 4. This relatively heavy deposit was probably due to flow turbulence about these protrusions as well as the inability to completely polish such areas because of mechanical interference. A tabulation of local activity for all three heaters is shown in Table 4 and a plot of the activity concentration along each cartridge heater is shown in Fig. 15.

The third heater indicated as much or more Cl^{36} activity as after Test No. 4. While the several areas which had been polished did produce "valleys" in the activity plot, these local reductions were not as great as would be expected from the reduction seen on the first two heaters. Comparison of the activity plot of the third heater with the activity plots from earlier tests indicated only minor differences. The similarity of the activity plots, plus the fact that polished surfaces still contained pitted areas similar to the previously unpolished area, indicated that pitting has a more pronounced effect on chloride deposition than general surface roughness. After the completion of the activity surveys, the three cartridge heaters were cleaned to remove as much of the Cl^{36} as possible.

Results of cleaning the first heater, which was clad with Type 347 stainless steel and which had been used several times previous to Test No. 6, are as follows:

- a) Washing in a 1% HNO_3 solution and scrubbing with a steel brush removed $4.7 \times 10^{-4} \mu\text{c}$;

Table 4 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 6*

Angle from reference, °	Distance from inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
<u>Upstream or first heater (downflow)</u>													
0	-	127	177	142	149	143	123	153	149	125	129	121	203
90	-	120	155	148	139	118	171	125	107	108	101	123	160
180	-	110	111	140	99	112	303	132	116	110	110	107	172
270	-	110	126	101	155	123	194	118	151	126	115	106	196
<u>Second heater (upflow)</u>													
0	-	153	103	120	113	112	104	248	105	100	105	130	108
90	-	136	130	95	99	99	110	125	105	120	106	107	108
180	-	164	117	112	98	110	116	166	109	94	118	105	105
270	-	185	119	92	109	117	106	143	113	110	105	126	85
<u>Downstream or third heater (downflow)</u>													
0	-	116	151	135	162	203**	303	223	190	221	262***	284	315
90	-	150	172	158	159	213	219	190	178	191	198	197	321
180	-	134	185	240	179	245	212	215	184	256	288	315	361
270	-	126	170	170	154	212	280	222	280	246	295	315	338

* Background = 85-110 counts.

** This column of readings read 155, 168, 145 and 153 after washing, scrubbing and soaking in 1% HNO₃.

*** This column of readings read 157, 154, 185 and 161 after washing, scrubbing and soaking in 1% HNO₃.

- b) soaking in 1% HNO_3 solution for two days removed an additional $4.3 \times 10^{-4} \mu\text{c}$;
- c) polishing to remove activity which still remained resulted in the removal of an additional $1.3 \times 10^{-4} \mu\text{c}$;
- d) the total activity removed from the first cartridge heater was $10.3 \times 10^{-4} \mu\text{c}$.

A similar washing and soaking procedure was applied to the second heater, which was also clad with Type 347 stainless steel and which had also been used several times previous to Test No. 6. Washing and scrubbing with 1% HNO_3 solution removed $4.0 \times 10^{-4} \mu\text{c}$, and soaking in the HNO_3 solution for two days removed $3.6 \times 10^{-4} \mu\text{c}$. No detectable activity remained after soaking, making the total activity removed equal to $7.6 \times 10^{-4} \mu\text{c}$.

The third heater, which was clad in Inconel and had previously suffered severe pitting, exhibited considerably more activity than either of the two upstream heaters. A two-step cleaning procedure similar to that applied to the other heaters removed 39.7×10^{-4} and $227 \times 10^{-4} \mu\text{c}$, respectively, from this heater, with some activity still remaining on the heater surface. Test results showed that this heater had collected a greater amount of Cl^{36} with each succeeding test and that removal of the resultant activity had become more difficult with each test. Apparently, a large fraction of the Cl^{36} became permanently imbedded in the pits, possibly after reacting with the surface constituents. In addition, the pits appeared to be more numerous and greater in depth after each test.

It was again noted that the total amount of chlorides collected by the heaters was only a small fraction of the amount passed over their

surfaces during the respective test. The first heater collected about 0.093%, the second about 0.069%, and the third, which was pitted badly, about 2.4% of the total chlorides which passed over the heaters.

5. Discussion of Test Results

Several types of deposition behavior became more apparent as the data for Test No. 6 were examined. These data, as well as earlier data, indicated that:

- a) Deposition increases sharply with increased surface roughness;
- b) collection of chlorides from steam may be substantially accelerated where surface pitting is present, even if a large percentage of the surface has a very smooth finish;
- c) chlorides tend to adhere quite tightly to very rough surfaces, and especially to pitted surfaces.

F. Chlorine-36 Test No. 7 - High Temperature Test

1. Test Objectives

Test No. 7 was designed to investigate the effect of high steam temperatures and/or high surface temperatures on chloride deposition.

2. Test Conditions

In order to obtain higher steam temperatures at the loop outlet (last cartridge heater), lower steam velocities were used; also, in order to obtain higher steam temperatures at the loop inlet (higher than had been investigated earlier), the preheater tube-side flow was set to obtain as much superheat (shell-side) as was possible. This operation resulted in entrance steam temperatures (at the first cartridge heater) between 590 and 600°F and exit steam temperatures (at the third heater) between

875 and 890°F. Cartridge heater surface temperatures varied between approximately 700°F at the inlet to 1050°F at the outlet. The steam flow rate was approximately 115 lb/hr. This flow rate resulted in velocities between 10 ft/sec (inlet) and 15 ft/sec (outlet) over the cartridge heater surfaces. The earlier tests had been operated with flow rates between 600 and 800 lb/hr, with the resultant higher steam velocities.

The surface finishes applied to the three cartridge heater (Type 347 SS) surfaces for this test were generally the finest to date. All exposed surfaces on the first heater were polished to a very bright and smooth finish using 600-grit emery paper, except for one section. The finish on the one section was left unpolished and was typical of as-received tubing finish except for being oxidized to a completely black condition by previous operation in steam. The second heater was oxidized slightly after polishing, to a straw-colored appearance, by heating in air before testing. The third heater was inserted into the loop in a recently polished, very bright condition. In general, only a minor number of pits or scratches were present on the polished areas before testing. The badly pitted heater was not used.

3. Test Procedure

As was done during earlier tests, the cartridge heaters were placed in the loop, steam was admitted at the desired rate, and then the preheater and conditioning heater were placed in operation. After superheated steam conditions had been obtained throughout the cartridge heater section of the loop, the NaCl³⁶ solution charging system was placed in operation and the injection of only distilled water was started. As

usual, this operation resulted in adding some Cl^{36} to the loop, as the injection piping was contaminated from previous tests.

The 520 to 525°F distilled water, and later the NaCl^{36} solution, was injected into steam which had been superheated to approximately 600°F by the conditioning heater located upstream from the injection point. This resulted in a flashing process which produced bulk conditions of slightly superheated steam (the saturation temperature was 530 to 532°F).

After equilibrium conditions had been established throughout the loop, 3 μc of NaCl^{36} were diluted to form four gallons of solution which was added to the charging tank. This solution was injected into the loop over a three-hour period and was followed by the usual 90-minute system distilled water cleaning process. The loop was then shut down, the cartridge heaters were removed, and the loop restarted. The preheater assembly was not removed. After restarting the loop, steam conditions comparable to those used during the test were re-attained and operation continued for several days. The injection pump using distilled water was operated for approximately two hours after restarting.

4. Test Results

The test data were obtained by collecting condensate samples of the loop effluent before, during, and after testing, by scanning the three cartridge heaters with a scintillation detector, and by counting all Cl^{36} which could be removed from the cartridge heaters by washing, scrubbing (steel brush), and soaking in 1% HNO_3 .

A plot of the effluent activity obtained during Test No. 7 is shown in Fig. 16. Surprisingly, this concentration did not vary greatly

from earlier tests when much larger steam flow rates were used. It had been noted that the effluent concentration in all tests seldom exceeds $1 \times 10^{-6} \mu\text{c/ml}$,* irrespective of injection rate-steam flow ratios. Unfortunately, this resulted in only about $0.2 \mu\text{c}$ of Cl^{36} being passed over the cartridge heaters before their removal. However, a high concentration was evident during several days of post-test operation and during this period the remaining $2.8 \mu\text{c}$ were exhausted to the bay.

By examining Fig. 16, it may be seen that some of the activity (between the hours 1000 and 1200) was a result of the injection system contamination from earlier tests. Then, after the $3.0 \mu\text{c}$ were added to the charging tank and Cl^{36} injection started, the effluent activity climbed to about $1 \times 10^{-6} \mu\text{c/ml}$ at the end of three hours of operation. After the charging tank was emptied of NaCl^{36} solution and distilled water was added, the effluent activity dropped slowly.

Following removal of the heaters and restarting the loop, the effluent concentration initially jumped to $100 \times 10^{-6} \mu\text{c/ml}$ which was more than a 100-fold increase over pre-shutdown values. As the concentration in previous tests had never been higher than $40 \times 10^{-6} \mu\text{c/ml}$, it was concluded that the loop piping had collected a major portion of the Cl^{36} during this low velocity, high temperature test. Concentrations between 0.7×10^{-6} and $1.0 \times 10^{-6} \mu\text{c/ml}$ persisted for several hours and operation with superheated steam for five additional days was required to remove all detectable activity from the loop.

* Except immediately after a startup when wet steam tends to flush the loop and in certain later tests.

A tabulation of heater surface activity readings for Test No. 7 is given in Table 5. This tabulation of total counts obtained during five-minute periods indicates little or no detectable activity on most heater surface areas. As the background normally ran between 79 and 110 counts in five minutes, only the readings with greater than 110 counts indicate detectable activity. A later cleaning process confirmed the presence of only a minute amount of Cl^{36} on each of these heaters.

A plot of the averages for each set of four readings, taken at 0, 90, 180, and 270 degrees, minus the average background, is shown on Fig. 17. It can be seen from this plot, as well as from Table 5, that the first heater collected the most chlorides, with a major portion of the deposition occurring on the unpolished, relatively rough, oxidized portion of the heater; the recently polished, bright area on this heater showed very little deposition.

The second heater, which had been polished and then oxidized slightly in air by heating to 900 to 1100°F also collected a small amount, with deposition occurring primarily about spacers.

The third heater which had been polished just prior to insertion in the steam loop, collected an amount similar to the second heater.

Each of the three heaters was then washed and scrubbed with a fine steel brush in 1% HNO_3 , soaked in a 1% HNO_3 bath for two days, and then scrubbed again. The first washing removed 1.6×10^{-4} , 2.6×10^{-4} , and 1.3×10^{-4} μc from the first through third heaters, respectively. The soaking removed 1.3×10^{-4} , 1.2×10^{-4} , and 0.7×10^{-4} μc , respectively. The two cleaning processes removed totals of 2.9×10^{-4} , 3.8×10^{-4} , and 2.0×10^{-4} μc from the three heaters.

Table 5 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 7*

Angle from reference, °	Distance from inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
<u>Upstream or first heater (downflow)</u>													
0	-	94	122	124	115	96	118	109	107	103	99	96	123
90	-	127	121	128	104	144	110	114	111	111	110	103	120
180	-	113	116	108	121	124	97	91	97	102	100	85	119
270	-	112	109	123	101	117	108	117	100	102	109	95	115
<u>Second heater (upflow)</u>													
0	-	120	101	-	83	-	106	95	-	87	-	88	-
90	-	169	85	-	95	-	93	110	-	102	-	93	-
180	-	147	108	-	86	-	118	178	-	114	-	105	-
270	-	135	105	-	101	-	85	122	-	92	-	108	-
<u>Downstream or third heater (downflow)</u>													
0	-	91	99	103	83	105	97	108	103	90	100	89	99
90	-	106	97	104	115	105	112	94	111	89	102	83	105
180	-	93	90	104	94	86	99	114	113	91	109	104	113
270	-	-	98	103	90	79	128	96	80	100	87	92	119

* Background = 79-110 counts.

While only small and comparable amounts of Cl^{36} were deposited on each heater, it should be remembered that less chlorides were passed over these heaters before their removal from the test loop than during any previous test. Only about 0.20 μc had left the loop during the test proper. Of the 0.20 μc , the first heater collected about 0.14%, the second 0.19%, and the third about 0.10%. The percentages were approximately twice the values noted after Test No. 6.

5. Discussion of Test Results

Because the amounts of Cl^{36} removed from the heaters were very small, quantitative conclusions concerning the test results may be suspect. However, it appeared that no significant temperature effect occurred and that surface roughnesses largely controlled the degree of deposition. The removal of more Cl^{36} from the second heater, which exhibited very smooth but oxidized surfaces, may indicate an ease of removal rather than greater collection. This belief is supported by the plot of average activity levels as shown in Fig. 17. This plot indicates a greater Cl^{36} deposition on the first heater which had a section of tubing with a relatively rough surface finish. This roughness would restrict removal of any chlorides as the deposition presumably occurs primarily in scratches, pits, etc.

Since the data from this test show that between 0.10 and 0.19% of the Cl^{36} which had been passed over the heaters was deposited on the heaters, some effect of the lower steam velocities is indicated. Earlier tests with velocities in the 50-60 ft/sec range resulted in collection of only 0.02 to 0.05%, where comparable surface finishes were used.

G. Chlorine-36 Test No. 8 - Repeat of High-Temperature, Low-Flow Test

1. Test Objectives

Test No. 8 was generally a repeat of Test No. 7. This earlier test was repeated since analysis of the data indicated that the cartridge heaters had been exposed to only about 0.2 μc of Cl^{36} during Test No. 7 and this exposure was not considered sufficient to produce activity levels which result in an acceptable counting accuracy. Too, a set of new Inconel-clad cartridge heaters had been prepared for loop testing and a repeat test seemed in order to produce data for comparison with earlier tests which used heaters that had been subjected to numerous acid baths and other cleaning procedures.

2. Test Conditions

The conditions for Test No. 8 approximated those of Test No. 7 very closely. However, as is explained below in Section 3, problems related to power supply equipment resulted in several temperature transients.

For the first time, all three cartridge heaters were clad in the same material (Inconel), had the same operating history (in this case all were new), and had identical surface finishes at the start of the test. However, after completion of the test it was noted that, because of differences in steam and surface temperatures, the degree of surface oxidization varied on the three heaters. The first heater had a straw-colored, light-reflecting surface, the second heater had a light brown surface finish and the third heater had a dark brown surface finish. This observation is pertinent since the degree of surface oxidization appears to affect the detectable activity level which results from collection and/or retention of the Chlorine-36.

The general test conditions, such as temperatures, pressures and velocities, were almost identical to those of Test No. 7. These conditions were such that initially slightly superheated steam at about 900 psia was passed over three cartridge heaters whose surface temperatures ranged from 600 to 1100°F. The steam was heated to about 850 degrees by these same heaters. Steam velocities ranged from 9 to 14 ft/sec.

The initial cartridge heater surface finishes were uniform and identical. The entire surface of each heater had been polished circumferentially with 600-grit emery paper. When the heaters were inserted into the loop they exhibited a uniformly bright and smooth appearance and were completely free of scratches and other surface defects.

3. Test Procedure

The test procedure was similar to that used for Test No. 7 except that the cartridge heaters were left in the loop much longer, and hence exposed to about 6 to 7 times as much NaCl^{36} . Also, two inadvertently large temperature transients occurred due to temporary losses of cartridge heater power. For the first time, the cartridge heaters were left in the loop overnight, even though all $3.0 \mu\text{c}$ of NaCl^{36} were injected during the first few hours of testing. As has been mentioned previously, irrespective of injection procedure, it required many hours to sweep the NaCl^{36} through the loop and hence over the cartridge heaters.

Data from previous tests had indicated that the test loop characteristics tend to always produce an exit steam activity concentration of about $1 \times 10^{-6} \mu\text{c/ml}$ until loop components near the injection point become sub-saturated with Cl^{36} . During the equilibrium activity concentration operation, the exhaust rate of Cl^{36} from the loop was almost

exactly proportional to the steam flow rate, under absolutely steady-state operation. However, it was found that any temperature, pressure, or flow rate change would produce increased concentrations at the loop exit. Large transients produced concentrations several times greater than the original steady-state value.

The general test operation followed the usual sequence. The three heaters were placed in the hot loop, steam flow was then initiated and injection of distilled water by the solution pump was started. After about one hour of stabilizing operation, the 3.0 μc of NaCl^{36} (diluted to about 12 liters) were added to the charging tank. Approximately two hours were required to inject the solution into the steam during this test. After the injection had been completed, distilled water was added to the charging tank three times, in 1-gal units. This latter procedure was typical of post-test injection system purging operation. After the third gallon of distilled water had been added to the loop steam, an additional 20-gal of distilled water were added to the tank so that operation could be continued throughout the night. This operation continued until the next morning, at which time the injection system was taken out of operation, the loop shut down, and the three cartridge heaters removed for scanning. Wet steam was then passed through the loop for several days in an attempt to remove the remaining Cl^{36} from loop surfaces.

During Test No. 8 and for the first time, two small (about 1/2 in. by 2 in. by 1/16 in.) stainless steel, oppositely charged plates were exposed to the contaminated steam flow. These plates were positioned downstream from the third heater and experienced temperatures in the

825 to 875°F range. A 110-volt potential difference was applied to the two plates from a small rectifier unit. Power was supplied to the parallel plate electrodes after the initial two hours of operation and was continued until the cartridge heaters were removed.

4. Test Results

The test data were obtained by taking loop effluent samples during and after the test, by scanning the three cartridge heaters for Cl^{36} after testing, and by counting all removable Cl^{36} on the three heaters.

A plot of the loop effluent activity is shown in Fig. 18. It can be seen from this plot that the exit concentration reached $1 \times 10^{-6} \mu\text{c/ml}$ very quickly, increased to much larger values several times because of loop transients, and then returned to a relatively steady-state value of about $1 \times 10^{-6} \mu\text{c/ml}$ until the test was terminated early the following day. Approximately 1.3 of the $3.0 \mu\text{c}$ flowed over the heaters and left the loop during the entire test. Because of the several rapid power changes and the relatively small number of effluent samples which could be obtained during the test, the plot of effluent activity may not exactly coincide with actual loop operation.

A tabulation of the data obtained by scanning the cartridge heaters is given in Table 6. It can be seen that the greatest average activity level was associated with the second heater and an intermediate level with the third heater. No reason is known for this pattern. Also, if the position of the maximum activity value in each set of four circumferential readings is noted (see underscored numbers), no discernible pattern is found. Any such pattern would possibly indicate improper positioning within the test loop piping, etc.

Table 6 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 8*

Angle from reference, °	Distance from inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
Upstream or first heater (downflow)													
0	-	104	110	<u>120</u>	<u>122</u>	<u>127</u>	193	103	113	121	109	104	148
90	-	114	112	107	112	115	<u>214</u>	120	134	125	117	134	151
180	-	<u>124**</u>	101	107	97	106	<u>177</u>	<u>123</u>	<u>145</u>	<u>127</u>	<u>148</u>	<u>140</u>	<u>195</u>
270	-	98	<u>123</u>	106	99	116	177	122	<u>106</u>	109	115	95	160
Second heater (upflow)													
0	-	119	<u>148</u>	136	131	125	147	<u>159</u>	<u>162</u>	118	138	<u>137</u>	132
90	-	137	<u>217</u>	135	<u>162</u>	143	<u>161</u>	<u>159</u>	<u>142</u>	<u>159</u>	130	<u>132</u>	127
180	-	<u>154</u>	136	127	<u>139</u>	145	<u>139</u>	<u>147</u>	133	<u>153</u>	<u>143</u>	134	<u>139</u>
270	-	<u>137</u>	131	<u>140</u>	134	<u>168</u>	148	<u>159</u>	157	126	<u>135</u>	132	<u>137</u>
Downstream or third heater (downflow)													
0	-	124	149	<u>164</u>	<u>141</u>	114	134	<u>156</u>	113	122	124	122	113
90	-	114	<u>171</u>	159	132	<u>134</u>	120	<u>139</u>	<u>144</u>	124	<u>135</u>	<u>149</u>	109
180	-	148	157	151	141	<u>128</u>	122	142	<u>128</u>	<u>128</u>	109	<u>136</u>	<u>133</u>
270	-	<u>153</u>	147	147	132	133	<u>148</u>	131	110	121	126	131	122

* Background = 88-120 counts.

** Note that each maximum circumferential reading is underscored.

The difference between the average of each set of four readings and an "average" background value is plotted in Fig. 19. This plot also indicates that the second heater collected the greatest activity. Once again, the effect of protrusions on collection is apparent. From this plot it appeared that the collection rate increased with surface oxidation, except for the third heater which was the most discolored and yet gave a lower average count.

The relative amounts of Cl^{36} removed from the three cartridge heaters did not agree with the apparent relative surface activity. An overnight soaking in 1% HNO_3 and a later scrubbing with a soft fiber brush removed $7.2 \times 10^{-4} \mu\text{c}$ from the first heater, $4.3 \times 10^{-4} \mu\text{c}$ from the second heater and $11.4 \times 10^{-4} \mu\text{c}$ from the third. This was surprising since the second heater, which exhibited the highest apparent surface activity, yielded the least Cl^{36} removed by soaking and scrubbing. While the scanning was not intended to provide quantitative data, it was surprising to see what appeared to be a direct conflict between two procedures. However, it is possible that several factors may at times be cumulative and hence produce what appears to be conflicting results. False indications may result from the activity profile plots because:

- a) The scanning plots are based on an average of only four unweighted readings. Many more readings should be taken before any quantitative interpretation would be possible.
- b) Heavy films of oxidation products may affect the activity levels which are noted by scanning of the heater surfaces, because of self-shielding.

- c) The Cl^{36} may be more easily removed from one heater surface than another, particularly where varying degrees of oxidation have occurred.

Because of these problems, plus others which arise from attempting to analyze data based on such minute amounts of material, indications of different rates of deposition may only be correctly interpretable where large differences were present. However, data which were based on the amount of Cl^{36} which was removed are believed to be more reliable than comparative appearance of the activity profile plots. This belief was supported by the ability to remove a large fraction of the Cl^{36} from a surface by soaking, etc., and then note a considerable reduction or complete absence in activity by a second careful scanning of specific areas.

The removal of 7.2×10^{-4} , 4.3×10^{-4} , and 11.4×10^{-4} μc from the heaters indicated that 0.055%, 0.033%, and 0.088% of the Cl^{36} which was passed over the heaters was collected by their surfaces. This resulted in a total of 0.18% being collected during Test No. 8; during Test No. 7, a much larger fraction (about 0.43%) had been collected. Since the heaters were exposed not only to considerably more Cl^{36} but to a much longer period of operation during Test No. 8, it is believed that the average net collection rate during this extended operation was lower than was experienced during the earlier short-term tests because of losses to the steam.

The Cl^{36} being used in the tests normally had a specific activity of at least 0.4 millicuries per gram or 2.5×10^{-3} grams of chlorine per microcurie. It is interesting to note that where as little as 5×10^{-4} μc

had been deposited on a 2-ft-long, 3/4-in.-OD heater, the activity could be plotted with some accuracy and a general indication of the more heavily contaminated areas was obtainable. As 5×10^{-4} μc consists of only about 1.3×10^{-6} gm of chlorine, the techniques being used enabled the detection of extremely small amounts of material. Even smaller amounts were detectable after chemical removal from the heater surfaces or when higher specific activity material was used.

The charged plate assembly mentioned above was removed from the loop along with the heaters. The two 1/2-in. x 2-in. x 1/16-in. Type 304 stainless steel plates were then detached from the assembly and each of the four sides monitored for Cl^{36} activity. Both sides of the positively charged plate exhibited easily detectable and similar levels (11 and 13 cpm above background) of activity. A similar counting procedure indicated no activity on one side of the other plate and only a barely detectable amount on the second side. Although a charge effect appeared to be indicated, when this experiment was attempted during later tests, contradictory results were obtained.

5. Discussion of Test Results

The data obtained during Test No. 8 indicated several possibilities concerning the deposition of chlorides from steam. These indications were:

- a) The rate of deposition may decrease with operation and hence the accumulation of chlorides is probably not linear with time for a specific set of conditions. This supposition is based on the reduced percentage collection seen during Test No. 8, as compared to Test No. 7;

- b) The chlorine in the high-temperature steam may possibly exist as charged particles, either as ions or small masses of solids. As charged particles, the chlorides may be attracted to specific surface areas. Where, heretofore, the relationship between Cl^{36} collection and pitted areas was believed to be purely physical (i.e., because of roughness and pockets) the data for Test No. 8 indicated that some electrical attraction may exist. It is possible that the chloride particles being transported by the dry steam may become charged during passage over metal surfaces and/or that specific areas in or about pits, particularly about corrosion product peaks, may become charged after suffering a loss of electrons in the same process.

An attempt was made in Test No. 9 to confirm an electrostatic effect on deposition of chloride ions but it was unsuccessful.

H. Chlorine-36 Test No. 9 - Special Surface Conditions Test

1. Test Objectives

The objectives of Test No. 9 were to:

- a) Expose oppositely charged electrodes to the contaminated steam, in order to detect whether Cl^{36} deposition is affected by electrical charge.
- b) Expose a heater which contained nine thin spacer rings which were brazed to the heater surface.
- c) Expose a cartridge heater whose clad surface contained numerous corrosion pits as well as laboratory-produced surface defects.

The charged electrodes were used in an attempt to check data from an earlier test (Test No. 8) which indicated a preference of the Cl^{36} for

the positively charged plate. The heater which contained the brazed spacer rings was exposed to the contaminated steam in an attempt to evaluate the effect of such protuberances on Cl^{36} deposition. The pitted and marred heater surface was exposed to obtain a specimen for autoradiographic study and to help determine the relative importance of several different types of surface defects.

2. Test Specimens

A heater, which had become pitted from earlier use, was selected as the one to be exposed for autoradiographic purposes. During the earlier tests, the pits on this heater had shown Cl^{36} deposits in certain areas which resulted in high counting rates (about 200 cpm). In addition to the corrosion pits, several mechanical and chemical faults were intentionally applied to the clad of this heater for comparison purposes. Nine mechanical indentations, nine aqua regia plus HF etched areas, and two cutting tool-applied cross marks were formed on the surface adjacent to a heavily pitted area. The nine mechanical pits were approximately 1/64-in. in diameter and 10 to 15 mils deep; the chemical "pits" were between 1/16 and 1/8-in. in diameter, and had a depth of no more than 1 to 2 mils maximum; the cross marks were about 1/2-in. in length and had a depth of about 10 mils; and the existing corrosion pits penetrated to a depth of 2 to 3 mils.

The several types of surface defects were used in an attempt to determine if any pit-sized physical conditions, such as the relatively smooth walls of mechanically produced pits versus the porous surface of chemically etched pits, would affect the deposition and retention of NaCl^{36} .

In addition, the man-made defects could be placed in a specific pattern which would simplify the association of exposed areas on X-ray film with the initial surface conditions. The numerous and randomly spaced corrosion pits had presented an almost impossible location problem in this respect.

During Test No. 9, the pitted heater was placed in the upstream position, a heavily oxidized heater was placed in the second position, and the brazed-ring heater was placed in the downstream position. The brazed-ring heater was covered with a heavy oxidization product film from previous exposure to the high temperature oxygenated steam.

To determine whether an electric charge would have any effect on the NaCl^{36} deposition behavior, a three-plate electrode test assembly was installed in the loop. Two of the plates were charged to a potential of ± 180 volts d-c from ground (in the loop piping). The third plate was not charged and was simply suspended in the flowing steam atmosphere as a control specimen. The three plates were positioned side-by-side in an upflow section of the loop, where they were exposed to 825°F superheated steam (about 300°F of superheat).

Because of materials problems, the neutral plate of the assembly was removed shortly after the initial installation of the assembly. A photograph of the electrode assembly (with the neutral plate removed) after completion of the test is shown in Fig. 20.

3. Test Conditions

The conditions during Test No. 9 were almost identical to those of Tests No. 7 and No. 8 except that the steam flow rate was slightly higher, yielding steam and clad surface temperatures which were slightly

lower than during the earlier tests. The inlet saturated steam was heated to approximately 550°F (about 18°F of superheat) before being passed over the first test cartridge heater. Passage over the three test heaters increased steam temperature about 275°F, to about 825°F. The heater clad surface temperatures were 600 to 1000°F, steam velocities were between 10 and 15 ft/sec, and the loop pressures were between 880 and 920 psia.

The conditions listed above were maintained throughout the test except for two transients which resulted from loss of prime to the NaCl³⁶ charging pump. These losses resulted from the presence of air bubbles in the solution being injected and were unavoidable because the NaCl³⁶ solution was being added to an empty (or almost empty) tank while the pump was operating. Once the addition was completed, a test was generally continued irrespective of minor interruptions, rather than experience numerous starts before completion of a given test.

The loss of charging system flow resulted in almost instantaneous boiling of the solution in the NaCl³⁶ charging system heater (concentric-tube, steam-heated). This local production of steam rapidly forced the downstream, slightly subcooled leg of solution into the loop. Under the low-steam flow (in the loop) conditions, it was believed that this slug of contaminated water passed through the loop preheater with only partial evaporation of the water taking place. At such times, the first or upstream cartridge heater, which is just downstream from the preheater, may have experienced cooling by wet steam. This occurrence allowed relatively large concentrations of NaCl³⁶ to escape from the preheater area, which normally acted as a constant concentration control unit, and the loop

effluent concentration was increased sharply for a short period thereafter. Any loop condition transient had a similar effect on loop effluent concentrations, with the magnitude of effluent concentration change being dependent on the type and magnitude of the transient.

4. Test Procedure

The test procedure was typical of earlier tests with the only significant differences being: The 3.0 μc of NaCl^{36} were injected at a slightly greater rate (less dilution); the charging tank was filled to a relatively high level after passing only one gallon of distilled water through the system in a cleaning operation; and the heaters were left in the loop overnight. This operation was almost identical to that of Test No. 8.

A pre-test problem arose when it was discovered that the plates of the electrode assembly were contacting each other as well as the loop pipe walls. The loop was shut down and the assembly removed for inspection. It was found that the insulating device, a piece of fired lava, which separated the two oppositely charged plates and supported a third neutral plate, had disintegrated. Apparently, the somewhat porous lava collected moisture internally during the loop startup and exploded when heat was added suddenly.

It was decided to continue the test with only the two oppositely charged plates suspended in a position downstream from the cartridge heaters. This arrangement was not very stable and it was noted the steam flow apparently caused the plates to flutter. Twice during the test, the plates apparently shorted together and caused a blowout of the low-current fuse in the d-c supply.

5. Test Results

The test data were obtained by collecting condensate samples of the loop effluent before, during, and after testing; by scanning the cartridge heaters and electrodes with a scintillation detector; and by exposing Kodak type KK X-ray film to high-activity areas on the pitted heater surface. No attempt was made to remove the activity from heater surfaces for quantitative evaluation.

A plot of test loop effluent activity is shown in Fig. 21. The effect of the transients caused by loop and injection pump startup can be readily seen on this figure for the time period of 1000 to 1500 o'clock. Even after a loop cleanup, and with no injection, each loop startup resulted in a minor amount of Cl^{36} being washed from loop surfaces and exhausted from the loop. This effect resulted from the wet steam flow which exists for a few minutes after startup. The startup activity peak at 1000 o'clock and rapid decrease in activity level thereafter, shown on Fig. 21, is characteristic of the transient loop startup behavior.

During the time interval from 1130 to about 1345 o'clock, the injection pump was adding distilled water from the NaCl^{36} tank to obtain loop equilibrium conditions. A rapid decrease in effluent concentration is apparent as the injection system piping was decontaminated by the distilled water. Following the establishment of equilibrium condition, the initiation of NaCl^{36} injection resulted in a rapid increase in effluent concentration. This concentration increased steadily until all of the NaCl^{36} solution had been exhausted from the tank and the system cleanup was in progress.

An interesting effect should be noted at time 1420 o'clock on Fig. 21. At this time, the effluent concentration level increased in the absence of any known loop transient except for a loss of injection flow. This loss of flow resulted in moderate loop temperature transients. Also, upon restarting the pump, the initial NaCl^{36} which was injected was apparently much cooler than the 520 to 525°F equilibrium temperature; thus, for the first few minutes after pump startup, the steam temperature may have been reduced to saturation and wet steam may have impinged on the first cartridge heater during such transients.

A tabulation of activity levels found by scanning the cartridge heaters is given in Table 7. The upstream heater, which was known to be pitted badly, showed the expected high level of activity due to Cl^{36} deposition. In addition, an area of very high activity was found on a section of the heater clad where only a moderate degree of pitting had occurred (see distance columns for 11 in. and 13 in. in Table 7); physically, this area is located on the lower center section of the heater, just below the thermocouple. Surprisingly, the surface areas which had been intentionally marred showed no greater activity than did adjacent, quite heavily pitted areas (see 180° readings, for column 5 in. through 9 in., upstream heater, Table 7).

In order to obtain a visual picture of the Cl^{36} deposition areas, autoradiographs were made of sections of the upstream cartridge heater. Both the area which exhibited unexpectedly high activity levels and the area which contained the mechanically and chemically produced surface defects, were wrapped in Kodak Type KK film and placed in a darkroom.

Table 7 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 9*

Angle from reference, °	Distance from inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
Upstream or first heater (downflow, pitted badly)													
0	-	122**	175	286	269	230	661	<u>1138</u>	264	215	199	204	225
90	-	<u>112</u>	<u>290</u>	<u>617</u>	219	276	302	<u>820</u>	218	191	173	152	232
180	-	113	<u>184</u>	<u>254</u>	172	182	192	217	209	<u>234</u>	<u>274</u>	<u>273</u>	271
270	-	111	239	427	<u>297</u>	<u>319</u>	<u>383</u>	548	<u>288</u>	<u>292</u>	<u>292</u>	<u>325</u>	<u>308</u>
Second heater (upflow, two finishes)													
0	-	<u>162</u>	<u>118</u>	101	<u>108</u>	113	106	119	137	125	111	136	112
90	-	<u>121</u>	<u>104</u>	<u>119</u>	<u>108</u>	123	94	132	118	<u>136</u>	130	125	115
180	-	<u>134</u>	116	<u>113</u>	<u>106</u>	<u>125</u>	<u>121</u>	<u>154</u>	134	<u>128</u>	<u>150</u>	<u>138</u>	<u>129</u>
270	-	<u>138</u>	105	104	102	<u>96</u>	104	121	<u>142</u>	125	142	126	111
Downstream or third heater (downflow, brazed-joint heater)													
0	-	116	129	127	<u>146</u>	<u>128</u>	<u>138</u>	111	131	131	114	124	114
90	-	<u>129</u>	<u>138</u>	123	<u>124</u>	<u>128</u>	<u>128</u>	<u>135</u>	123	123	<u>132</u>	124	<u>135</u>
180	-	<u>115</u>	127	<u>150</u>	110	<u>106</u>	120	109	<u>136</u>	<u>136</u>	129	<u>129</u>	105
270	-	114	128	126	97	88	106	104	112	112	96	113	110

* Background = 85-108 counts.

** Maximum within each group of four circumferential readings is underscored.

After 13 days of exposure, the film was removed and developed. The exposed film clearly outlined the unexpected area of high activity but did not indicate any relationship between surface defects and deposition except at the chemically-produced pits and about some corrosion pits. A print of the film which was placed over the former area is shown in Fig. 22a; a pattern of activity which indicates a condition of wetting and drying is clearly evident. Apparently, moisture in the wet steam, which passed through the preheater as a result of the injection pump flow transient, was deposited on the heater thermocouple and then evaporated on the adjacent surfaces of the heater. After the exact location of the active deposit had been determined by the autoradiographs, the pattern of loop corrosion product deposits was found by visual observation, using special lighting techniques. This visual pattern duplicated the autoradiograph pattern exactly, as indicated by Fig. 22b which is a photograph of the area.

Figure 23 contains a photograph and an autoradiograph of the surface area which contained the various types of defects. While the detail of the autoradiograph is not exceedingly sharp, it can be seen that some corrosion pits are characterized by small but intense activity areas, other corrosion pits by large, less dense activity areas, but that the much deeper mechanical defects are not visible, indicating no activity.

The procedure of exposing Kodak Type KK industrial X-ray film to cartridge heater surfaces which had been contaminated with the Cl^{36} in high pressure steam was repeated later. This latter set of film was

exposed for more than one month, while the earlier set had been exposed for less than two weeks. The latter set of negatives duplicated earlier indications of large deposits of Cl^{36} in or about pits and on areas which had experienced wet steam flow. The same pitted, Inconel-clad cartridge heater which had been used earlier was used as the source for these recent autoradiographs also.

An autoradiograph of an area of the heater which had been subjected to "run-down" of water from saturated steam is shown in Fig. 24a. The effect of this wetting and drying can be seen by the two longitudinal "trails" of Cl^{36} . The effect on deposition of a thermocouple extension (1/16-in. OD) can be seen also; the sheathed thermocouple extension was pressed against the heater surface and influenced deposition by routing some of the moisture rundown. Star-like spots of heavy local contamination, which are typical of pitted surfaces, are visible also.

An autoradiograph of a more heavily pitted section of the heater is shown in Fig. 24b. No cloud-like deposits were found in this area as this downstream portion of the heater experienced only superheated steam flow. However, the results of a myriad of microscopically-sized heavy deposits of Cl^{36} are visible. In some areas, families of deposits, which would be associated with families of pits, are discernible. Rows of deposits, which could not be associated with any series of pits, indicate the influence of tube forming, marring during test assembly, or some other surface condition disturbance.

No undebatable theory as to why these deposits occur is available. Not all pits collected detectable amounts of the Cl^{36} and the size of the

exposed area on the film is not representative of the size of the suspected pit. Some very large pits do not appear to have collected any Cl^{36} . In addition, film indications of very large deposits of Cl^{36} may have been produced by an area of the heater which exhibited patches of very small pits. However, in all instances, the number of exposed spots on the film appeared to be proportional to the number of pits on the metal.

No visible deposit of any material was found on the lower section of the cartridge heater, which was cooled by superheated steam.* The beta particles which produced the pattern shown in Fig. 24b, emanated from invisible deposits of Cl^{36} . The compound or chemical form of the Cl^{36} is not known. Presumably it exists as either NaCl^{36} (the source), or as salts formed by the elements in the metal.

Table 7 also includes tabulation of the activity found on the other two downstream heaters. A plot of the average net count rate for all three heaters is shown in Fig. 25. The very high level on the first heater is apparent; this level resulted from Cl^{36} deposition in surface pits plus moisture rundown and evaporation on the heater clad surface. The second and third heaters exhibited much lower activity levels. This was somewhat surprising in that both had highly oxidized, relatively rough surfaces and the third heater contained numerous rings and rough brazed areas. Since the surface of the second heater was only moderately rough, this deposition behavior may be expected. However, the surprisingly low activity of the third heater is not understood. One explanation lies in the possibility that a combination of the heavy oxidization products

* The surface was oxidized to a dark brown condition.

film and prior collection of loop corrosion products in pits and rough areas may have limited the Cl^{36} deposition. Both of the heaters had operated for two weeks in 700-900°F steam, with surface temperatures between 900 and 1100°F, prior to exposure to the contaminated steam.

It is interesting to compare the activity plot of the second heater (Fig. 25) with a plot taken for the same heater after an earlier test. For Test No. 7, one-half of this heater had been polished, the other half left "as-received" (slightly oxidized) and the heater exposed to Cl^{36} . The activity plots obtained for both tests are quite similar as can be noted in Fig. 26.* Even though the heater was exposed to several times as much Cl^{36} in Test No. 9, because of longer exposure time, the activity levels were similar. However, the most significant point is the almost identical shape, rather than the magnitude, of the two plots.

A photograph of the electrode assembly which was used during Test Nos. 8 and 9 (taken after use and after removal of the neutral electrode) is shown in Fig. 20. The electrodes hung vertically downward in a 1-in. Schedule 80 pipe with steam flowing in an upward direction during the test. The holes for suspending the neutral electrode are visible near the lower ends of the plates. An attempt had been made to position all three plates as nearly parallel to each other as possible. The dark object just above the plates is a centering device which is attached to a MgO insulating bead. As mentioned earlier, similar and

* It was necessary to reverse the prior plot, with respect to upstream and downstream directions, as the heater had been exposed in different sections of the test loop.

barely detectable amounts of Cl^{36} were found on each of the four sides of the two electrode plates which were tested.

Since the three heaters were not cleaned, a quantitative determination of the amount of Cl^{36} which was collected was not possible. The pitted heater was reserved for autoradiographic purposes and the brazed-ring heater and standard heater were set aside for future test work.

6. Discussion of Test Results

Several pertinent observations were made when the data for Test No. 9 were examined. These were as follows:

- a) No effect was noted of charged plates on NaCl^{36} deposition from superheated steam. However, it can be hypothesized that, if the chlorine were in the ionic form (as Cl^-), then contact with a positive plate may convert the ions to gaseous chlorine which is swept away by the steam. Nevertheless, it was surprising that there was little or no activity on either plate, since all objects normally collect easily detectable amounts under the loop conditions.
- b) There was obvious agreement in deposition patterns on the same heater after two different tests. However, it was not apparent why the apparent deposition was not proportional to the amount of chlorides which were passed over the heater, under otherwise similar conditions. It does not appear possible that "saturation" deposition levels were approached this quickly.
- c) Autoradiographs of areas of the heaters clearly illustrated the effect of drying wet steam on the deposition of solids.

- d) The large mechanically-produced surface defects apparently had a minor effect on chloride deposition, when compared to the effect of roughness which resulted from corrosion pitting and chemical etching.

I. Chlorine-36 Test No. 10 - Use of MgCl_2^{36}

1. Test Objectives

The primary objective of Test No. 10 was to determine if MgCl_2 behaved similar to NaCl in flowing steam. The relative deposition rate from flowing superheated steam was the item of major interest. During all previous tests, the Cl^{36} was injected into the steam loop as a dilute solution of NaCl^{36} , with a pH of 7-8. This chemical form was used since the chlorides in the BONUS coolant are expected to exist primarily as NaCl, because the in-leakage from the sea water-cooled main condenser is expected to be the major source of chlorides and salt-forming cations. However, other chloride salts will exist and it was believed to be necessary to compare the behavior of additional salts. The probability that MgCl_2 would constitute the second largest source of chlorides, plus the somewhat unique behavior of MgCl_2 under drying conditions, prompted this selection.

2. Test Conditions

Test conditions approximated those used during Tests 7 through 9. The steam pressure ranged between 875 and 925 psia, the steam temperature from 550°F (18°F superheat) to 900°F, and steam velocities in the 8-12 ft/sec range. Low steam velocities were used in an attempt to obtain greater deposition.

Three 3/4-in.-OD, 24-in.-long Inconel-clad cartridge heaters were exposed to the superheated steam. Prior to testing, each heater had been

polished to a very smooth condition by using 600-grit emery paper. When the heaters were installed in the loop, each surface exhibited a bright, unoxidized, almost mirror-like finish. Since the three heaters operated in series, each successive heater experienced higher steam and surface temperatures, with the maximum surface temperature of the downstream heater ranging between 1100 and 1150°F.

3. Test Procedure

The test procedure was typical of earlier tests. The 3.0 μc of Cl^{36} , as 0.15 ml of 3.6N HCl^{36} , were neutralized by adding 0.045 gm of $\text{Mg}(\text{OH})_2$ to the acid after it had been diluted to about 1.5 ml. This solution of MgCl_2^{36} was then further diluted, to 9000 ml, before injection into the test loop. After the loop had been brought to equilibrium conditions, the MgCl_2^{36} was injected over a period of 170 minutes. After completion of the injection, distilled water was added to the charging tank and this relatively uncontaminated water was sprayed into the loop for almost three hours, after which the injection pump was stopped. Normal plant steam was then passed through the test loop under test conditions until the next morning. At that time, the loop was shut down and the three cartridge heaters removed for examination. The loop was then restarted and operated for several days for decontamination purposes.

The three cartridge heaters were scanned for radioactivity and later soaked for 24 hours in a 1% HNO_3 solution. This wash solution was processed in the usual manner, in order to collect the Cl^{36} , as AgCl^{36} , for counting and quantitative evaluation.

4. Test Results

The test data were obtained by collecting condensate samples of the loop effluent before, during, and after testing; by scanning the cartridge heaters and electrodes which were exposed to the contaminated steam flow; and by removing the chlorides from the heaters for quantitative evaluation of the deposition.

A plot of test effluent activity is shown in Fig. 27. This plot provided the most interesting data obtained during Test No. 10. Between 1000 and 1100 o'clock, the effect of loop startup was typified by the small amount of activity which appeared and then decreased toward a barely discernible value. Another increase in activity is seen between 1100 and 1200, after the injection pump was started. Both of these increases are typical of most tests; the former occurred as wet steam passed through the loop; the latter resulted from the small amount of Cl^{36} contained in the injection system from the previous test.

About one hour after the MgCl_2^{36} had been added to the injection system, the effluent activity rose to about 15 to 16×10^{-6} $\mu\text{c}/\text{ml}$. This was the greatest effluent concentration ever obtained under steady-state loop operation. This concentration was surprising since for all previous test conditions, except for the use of MgCl_2^{36} , the concentration had never exceeded 6×10^{-6} $\mu\text{c}/\text{ml}$.

Another surprising effect was noted when the MgCl_2^{36} solution was expended. Within one hour after completion of the injection, the effluent activity dropped to about 5×10^{-6} $\mu\text{c}/\text{ml}$. For similar earlier tests, this decrease occurred at a much reduced rate.

The plot of effluent activity indicated that the MgCl_2^{36} had passed directly through the loop, without the usual gradual removal of Cl^{36} by deposition on the preheater and other components, upon which the injected solution is dried. Graphical integration of the effluent activity plot revealed that 2.6 μc of the Cl^{36} , or 87%, had passed directly through the loop. Normally, only about 20 to 25% of the injected material exists during the test operation. It was evident that, barring some unknown test procedure difference, the MgCl_2^{36} had behaved quite differently than had earlier injections of NaCl^{36} .

There appears to be at least one logical reason for the difference in behavior between NaCl and MgCl_2 solutions which are reduced to dryness in superheated steam systems. This difference stems from the inability to reduce MgCl_2 solutions to MgCl_2 by addition of heat under ordinary conditions. Apparently, when evaporation of a solution of Mg^{++} and Cl^- ions is attempted, a precipitate of $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ appears, which, upon further heating, produces MgO (solid), HCl (gas) and H_2O (steam). This reaction apparently occurred in the steam loop and the Cl^{36} passed through the loop as HCl^{36} , not as MgCl_2^{36} . Since HCl is presumably a true, rather inert gas under superheated steam conditions, this reaction appears to have been responsible for the rapid transit of the Cl^{36} through the loop.

A tabulation of the activities found by scanning the three cartridge heaters is shown in Table 8. It can be seen from this tabulation that very little Cl^{36} was collected by these heaters. A graphic indication of the small degree of deposition can be seen in Fig. 28. Here, the averages of each set of four circumferential activity values is plotted

Table 8 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 10*

Angle from reference, °	Distance from steam inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
<u>Upstream or first heater (downflow)**</u>													
0	107	89	89	95	106	184	202	102	96	83	105	85	103
90	91	108	102	98	86	145	603	110	85	83	96	112	119
180	112	99	122	95	87	102	137	115	90	101	91	96	89
270	112	97	104	98	115	138	148	102	101	101	93	103	114
<u>Second heater (upflow)</u>													
0	-	92	96	102	115	97	94	98	93	98	91	106	112
90	-	110	104	108	105	102	94	100	99	112	106	110	91
180	-	112	90	96	91	106	100	73	102	91	80	127	108
270	-	96	97	95	104	94	108	95	83	82	98	93	86
<u>Downstream or third heater (downflow)</u>													
0	-	111	113	98	112	99	101	101	76	106	102	110	83
90	-	101	103	102	99	81	97	102	101	89	90	115	119
180	-	98	104	105	103	80	105	89	77	102	83	90	115
270	-	96	87	123	78	92	96	103	91	91	107	86	114

* Background = 95 counts.

** All heaters were Inconel and polished with 600-grit paper before test.

versus heater length. Only slightly detectable levels of activity are visible from this plot, except near the center of the first heater where, apparently, some moisture was dried about a thermocouple weld. The almost complete absence of Cl^{36} was further established when all removable chlorides were processed and counted. Totals of 3.3×10^{-4} , 0.86×10^{-4} and 1.2×10^{-4} μc were removed from the upstream to downstream heaters, respectively. These levels were the lowest ever found after any of the low-velocity, superheated steam tests. If the first heater deposition value is ignored, because of the presumed temporary wet steam flow, the amounts of Cl^{36} which were collected are about one-tenth to one-fourth those found after earlier, similar tests. It should also be noted that during this MgCl_2^{36} test, 2 to 3 times as much Cl^{36} was passed over the heaters, compared to Test No. 9, where similar conditions and the same cartridge heaters were used.

Presumably, the rapidity with which the Cl^{36} passed through the steam loop was indicative of the lessened tendency toward deposition on piping surfaces, etc., as well as on the cartridge heater surfaces. It was believed that, with all other steam conditions being identical, a high effluent concentration (activity level) was indicative of a low deposition situation within the loop.

No preferential deposition of the Cl^{36} on either of the charged plates was noted.

5. Discussion of Test Results

It is believed that, during Test No. 10, the use of MgCl_2^{36} resulted in a large percentage of the Cl^{36} acquiring the form of HCl^{36} , a

compound of gaseous nature when in 900 psig superheated steam. In addition, the decreased chloride deposition was believed to be a direct result of the HCl^{36} production.

If the drying process associated with the contaminated, originally subcooled water which was sprayed into the loop is examined, it can be seen that several chloride compounds will generally be produced and, where a high purity solution was used, it is not possible to ever produce one pure compound. For instance, if a solution of water, 0.1 ppm Mg^{++} , 0.1 ppm Na^+ and 0.4 ppm Cl^- were used, compounds of both $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ and NaCl would be formed (the $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ temporarily) as a droplet (or other body of liquid) by concentration through evaporation.

Under ordinary conditions, numerous chloride compounds will be formed, depending upon the relative amounts of the various ions and their solubilities at the conditions under which evaporation and concentration occurs. However, for solutions of high purity, it may be possible to intentionally increase the concentration of one material such that preferential formation of a specific compound will occur. This was, presumably, the situation during this MgCl_2^{36} test. Relatively large amounts of Mg^{++} , as $\text{Mg}(\text{OH})_2$, were added to a high purity, high pH water sample and later, upon concentration by evaporation in the loop, the chlorides achieved the form of $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ primarily, and then HCl . In the test, the Mg^{++} concentration exceeded the Cl^- concentration by at least 100% and the Na^+ concentration by about a factor of ten.

It now appears that the behavior of chlorides in steam systems may depend upon the chemical form which exists within the solution (boiler water), the relative amounts of contaminants and their relative solubilities,

and even the solution pH. Also, it appears generally impossible to carry over MgCl_2 in high-temperature steam systems, wherein the moisture containing MgCl_2 is thoroughly dried. Apparently, MgO and HCl are produced by the forced drying.

J. Chlorine- 36 Test No. 11 - Successive Additions of Various Solutions of Cl^{36}

1. Test Objectives

The objective of Test No. 11 was to substantiate several indications of chloride behavior seen during Test No. 10. Primarily, the test was designed to answer the question of whether HCl^{36} had actually been produced by drying the MgCl_2^{36} under loop conditions. With this primary objective in mind, the usual $3.0 \mu\text{c}$ of Cl^{36} were injected as $1.0 \mu\text{c}$ of NaCl^{36} , $1.0 \mu\text{c}$ of low-pH NaCl^{36} and $1.0 \mu\text{c}$ of HCl^{36} . Each microcurie was injected separately and 0.5 gal. of uncontaminated water was injected between each injection of Cl^{36} solution. If the assumptions stated above concerning Test No. 10 were correct, then the effluent activity plot should be similar to the plot for Test No. 9 during the NaCl^{36} injection, similar to a combination of No. 9 and 10 during the low pH NaCl^{36} injection, and similar to Test No. 10 during the HCl^{36} injection.

Negatively and positively charged electrodes were exposed to the contaminated steam for the fourth time.

2. Test Conditions

The test conditions and cartridge heater surfaces were identical to those used during Test No. 10. This information is given above in Section I-2.

3. Test Procedure

Except for the preparation and injection of the Cl^{36} solution, the test procedure was identical to that of Tests No. 9 and No. 10. During those tests, the $3.0 \mu\text{c}$ of Cl^{36} were diluted to 9000 ml and injected into the operating test loop in a continuous manner. The solution for use in Test No. 9 was obtained by taking steam plant condensate, adding the 4.2 ml of 0.003N HCl^{36} and then adding 60 ml of 0.2N NaOH . The procedure for preparation of the Cl^{36} solution for Test No. 10 is described above in Section I-3. The $3.0 \mu\text{c}$ of Cl^{36} (each microcurie as 0.05 ml of 3.6N HCl^{36}) used in Test No. 11 were prepared as follows:

- a) The first microcurie was prepared as was done for Test No. 9. No compensation was made for the different HCl^{36} normalities, etc. The final solution pH was 8.0.
- b) The second microcurie was prepared as was done during Test No. 9, except, after the preparation, the solution pH was reduced to 3.5 by the addition of HNO_3 .
- c) The third microcurie was prepared by simply adding the HCl^{36} to plant steam condensate. The pH was then found to be approximately 6.0. Therefore, the hydrogen ion concentration was about 1×10^{-6} equivalents per liter, the sodium ion content was estimated to be about 10 ppb or 0.5×10^{-6} equivalents per liter, and the chloride ion content about 60×10^{-6} equivalents per liter.

After the injection of each microcurie was completed, 0.5 gal. of plant condensate was added to the injection system tank and injected in

a rather ineffective attempt to decontaminate the injection system. After injection of the third microcurie, two 0.5 gal. additions of condensate were used and then the injection system pump was stopped. The loop was left in operation until the next morning when the cartridge heaters were removed.

4. Test Results

The test results definitely supported the data obtained from Test No. 10. It can be seen from the plot of loop effluent activity in Fig. 29 that the activity level of effluent related to the high pH NaCl^{36} solution increased rather slowly and appeared to be approaching an equilibrium value of $6.8 \times 10^{-6} \mu\text{c/ml}$. The level then decreased upon the addition of initially uncontaminated water. After addition of the 3.5 pH solution of NaCl^{36} to the charging tank, the level increased at a very rapid rate until the second batch of uncontaminated water was added to the tank. The addition of the HCl^{36} solution resulted in another marked increase in activity and it then appeared that the effluent activity concentration was approaching equilibrium at a value of approximately $16 \times 10^{-6} \mu\text{c/ml}$. It is interesting to note that the apparent equilibrium value of $6.8 \times 10^{-6} \mu\text{c/ml}$, associated with the NaCl^{36} injection agrees quite well with a value of about 5.5×10^{-6} which was attained during Test No. 9 which was a NaCl^{36} test. Also, the final apparent equilibrium value of $16 \times 10^{-6} \mu\text{c/ml}$, associated with the HCl^{36} injection, agrees with the value of 15 to 16×10^{-6} associated with the MgCl_2^{36} test, which presumably produced a large percentage of HCl^{36} (see Fig. 27). In addition, the peak value (apparently not close to equilibrium) associated with the

low pH NaCl^{36} solution injection was much higher than any level experienced during steady-state operation of any NaCl^{36} test. It therefore appears that Test No. 11 effluent concentration data reproduced earlier NaCl^{36} equilibrium concentration data, reproduced the earlier HCl^{36} data (from MgCl_2^{36}), and indicated a strong influence of the solution pH.

Although the several levels seen during Test No. 11 were not maintained for sufficient time to unquestionably ascertain the exact equilibrium values, it is believed that the data were sufficient for a general analysis. A test duration time about twice that used would probably have been required to obtain true equilibrium conditions. Such a test would have been very difficult to perform since it would have required over 20 hours of continuous effort by the test personnel.

When examining the plot given in Fig. 29, the time lag, between the addition of the first NaCl^{36} to the injection tank and the appearance of increased activity at the loop exit should be noted. This time lag is approximately constant for each addition and indicates that the injection system flow behavior is essentially identical for each addition while the amount and rate of effluent activity varied considerably with each addition.

A tabulation of activity level data obtained by scanning the three cartridge heaters after removal from the loop is shown in Table 9. Only a few areas provided readings which were discernible from background values. A plot of the average circumferential activity found on the three cartridge heaters is shown in Fig. 30. As during Test No. 10, the activity, and therefore Cl^{36} , levels were barely above background. In general, the activity appeared to exist primarily about spacers and thermocouples.

Table 9 TABULATION OF HEATER ACTIVITY, TOTAL COUNTS IN FIVE MINUTES-TEST NO. 11*

Angle from reference, °	Distance from steam inlet, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
<u>Upstream or first heater (downflow)</u>													
0	-	99	99	109	89	110	123	78	96	94	91	72	124
90	-	105	95	97	102	108	191	100	99	111	112	101	113
180	-	84	92	100	98	108	106	71	99	119	116	96	146
270	-	98	92	85	112	112	140	93	106	95	90	86	136
<u>Second heater (upflow)</u>													
0	-	170	98	98	103	101	98	97	99	104	105	92	96
90	-	179	89	85	112	80	88	81	108	111	98	103	93
180	-	170	91	96	86	109	85	101	96	103	75	86	96
270	-	119	81	104	91	101	99	118	92	105	87	73	87
<u>Downstream or third heater (downflow)</u>													
0	-	***	***	74	93	105	123	89	105	104	106	96	95
90	-	-	-	104	103	103	112	106	122	86	86	98	118
180	-	-	-	95	97	103	111	106	94	101	111	102	104
270	-	-	-	96	108	106	113	106	95	104	106	95	110

* Background = 90 counts.

** Not counted.

Soaking, washing, and brushing the three cartridge heaters in 1% HNO_3 removed $1.41 \times 10^{-4} \mu\text{c}$, $1.59 \times 10^{-4} \mu\text{c}$ and $0.3 \times 10^{-4} \mu\text{c}$ from the three heaters upstream to downstream, respectively. These values are comparable but generally lower than the amounts removed after Test No. 10. It was expected that these values would be greater than those for the earlier test because of the use of some NaCl^{36} , and because an increased deposition was indicated by the activity plots. However, where amounts of this magnitude are encountered, quantitative correlations were not possible; also, the first heater in Test No. 10 presumably experienced some wet steam flow which deposited a relatively large amount of Cl^{36} on that heater.

As during the previous test (No. 10), no activity was found on either of the stainless steel electrodes which had been charged to ± 110 volts d-c.

5. Discussion of Test Results

The data taken during Tests No. 10 and No. 11 indicated quite strongly that the chemical form of chlorides, in the moisture phase, may affect the deposition behavior of chlorides traveling through a steam-filled system. This is especially true where the chlorides may exist as HCl , as opposed to a salt such as NaCl .

The absence of Cl^{36} on either of the charged electrodes which were exposed to the contaminated steam was puzzling. Several voltages, etc., have produced no results in three of four tests. Therefore, it was then assumed that either no effect of electrically charged surfaces existed or that the negative plate repelled the Cl^{36} and the positive plate oxidized

the Cl^- to Cl_2 , which then left the loop as a gas and hence left no deposit on the positive plate.

K. Laboratory Carryover Tests

1. General

Several evaporation-to-dryness tests, using several different solutions of Cl^{36} , were conducted in an attempt to determine the relative amounts of Cl^{36} which remain in the solution residue versus the amounts which leave the boiling solution or drying residue with the steam. It was assumed that, except for a small amount of physical carryover, all Cl^{36} which left the solution had acquired the form of either HCl^{36} or Cl_2^{36} .

A sketch of the apparatus in the bench tests used is shown in Figs. 31 and 32. The apparatus shown in Fig. 31 was used initially; the apparatus shown in Fig. 32 was used during later tests (H and I) in an attempt to prevent loss of gaseous products during post-test equipment disassembly. A total of 0.0067 microcuries of Cl^{36} was used in each test.

After each solution was evaporated to dryness, the hotplate power was left on for 10 minutes; during this period the hotplate surface temperature was in the 700 to 800°F range and it is estimated that the residue temperature was in the 300 to 400°F range. Both condensate and residue were processed later to determine the amounts of Cl^{36} that were present. Any remaining sweeping steam and water were also processed. It was found that up to 5% of the original Cl^{36} migrated upstream to the sweeping steam source (water in flask).

2. High pH Solutions of NaCl^{36}

The NaCl^{36} solutions were obtained by diluting 1.0 μc of HCl^{36} , in the form of 0.15 ml of 0.44N HCl^{36} , to 3000 ml; withdrawing 20 ml of the

dilute HCl^{36} ; then diluting the withdrawn solution to 120 ml total with deionized water; and adding 2.0 ml of 0.25N NaOH. This produced a dilute solution of NaOH and NaCl^{36} with a pH of 10 to 11. This solution was added to a 250 ml Pyrex flask and evaporated on a hotplate. The steam produced by boiling this solution was passed through a 1/4-in. Type 304 stainless steel tube to a condensing-collecting beaker filled with deionized water.

The data for these tests are tabulated in Table 10. Tests No. A and F involved the use of high pH solutions of NaCl. Only a small amount of the Cl^{36} was recovered in the condensate collector during these tests. After the latter test, all of the Cl^{36} had been generally located, with 95% remaining as residue from the dried solution. Only about 71% of the Cl^{36} used during the first test was recovered; however, the remaining 29% was believed lost when the distilling flask broke during post-test heating. It is probable that the 4 to 5% recovered in the condensate had been carried over in moisture droplets.

3. Low pH Solutions of NaCl^{36}

The conditions related to the two tests described above were repeated during Test No. C, except that the solution pH had been reduced from 11.0 to 2.0 by the addition of HNO_3 prior to evaporation. Here the results were significantly different from those discussed above. The residue contained only about 18% of the original Cl^{36} while 15% was collected in the condensate. The remainder was believed lost during post-operation equipment disassembly, as gaseous HCl^{36} or Cl_2^{36} which either escaped the condensate collector or reacted with equipment surfaces. Either theory would require that the Cl^{36} exist primarily as a gaseous material,

Table 10 TABULATION OF DATA FOR LABORATORY BENCH-TYPE CARRYOVER TESTS

Test No.	Solution ⁽¹⁾	Solution vol. ml	Solution ion content, ppm				pH (Approx.)	% of original Cl ³⁶ in	
			Na ⁺	Mg ⁺⁺	H ⁺	Cl ⁻		Residue	Distillate
A	NaCl ³⁶	120	96	nil	1x10 ⁻⁸	0.14	11.0	67 ⁽²⁾	4
B	MgCl ₂ ³⁶	120	nil	0.22 ⁽⁷⁾	1x10 ⁻⁴	0.14	7.0	96	4
C	NaCl ³⁶	120	96	nil	10	0.14	2.0 ⁽³⁾	18	15
E ⁽⁴⁾	MgCl ₂ ³⁶	120	nil	0.22	1x10 ⁻¹	0.14	4.0	61	14
F ⁽⁵⁾	NaCl ³⁶	120	96	nil	1x10 ⁻⁸	0.14	10.5	95	5
G ⁽⁶⁾	MgCl ₂ ³⁶	120	nil	0.22	1x10 ⁻¹	0.14	4.0	53	5
H	MgCl ₂ ³⁶	120	nil	0.22	1x10 ⁻²	0.14	5.0	49	7
I	MgCl ₂ ³⁶	120	nil	0.22	10	0.14	2.0 ⁽³⁾	4	46

(1) Only 0.0067 microcurie used in each test.

(2) A large fraction of the residue was lost when the flask broke.

(3) The low pH was produced by the addition of HNO₃.

(4) Due to an error in the test procedure, Test No. D was not completed.

(5) Repeat of Test No. A.

(6) Repeat of Test No. E.

(7) Because of the preparation procedure employed, this value is questionable.

presumably as HCl^{36} or Cl_2^{36} . It should be remembered that only about 1.6×10^{-5} gm of Cl^{36} was used for each test. This would have produced only about 0.01 ml of gaseous HCl^{36} , or 0.005 ml of Cl_2^{36} .

4. Solutions of MgCl_2^{36}

Five separate tests were conducted with MgCl_2^{36} . After four of these tests were completed, between 4 and 61% of the original Cl^{36} remained in the residue. About 96% of the Cl^{36} was found in the residue of the fifth test; however, the results of this test should be disregarded as questions exist concerning the preparation of the MgCl_2^{36} .

During one of the four initial tests, a low pH solution of MgCl_2^{36} was used (Test I). After this test only 4% of the Cl^{36} remained in the residue. After a similar test, using NaCl^{36} , 18% of the Cl^{36} was found in the solution residue. These data indicate some contribution of the MgCl_2^{36} toward producing HCl^{36} or some other gaseous chlorine compound.

The procedure for the MgCl_2^{36} tests was identical to that of the NaCl tests, except for the production of MgCl_2^{36} . This compound (in solution) was produced by either the addition of dilute $\text{Mg}(\text{OH})_2$ solution, or the addition of MgO , to the HCl^{36} solution.

5. Repeat Tests

Two of the evaporation-to-dryness tests which have been described earlier were repeated in an attempt to develop a procedure whereby 100% of the Cl^{36} could be accounted for after each test; one of these tests used NaCl^{36} and the other MgCl_2^{36} . As in the previous tests, greater than 90% of the Cl^{36} remained in the residue when the high pH solution of NaCl^{36} was evaporated, while only about 10% of the Cl^{36} remained after

evaporation of a solution of MgCl_2^{36} . Again, it was noted that after the MgCl_2^{36} test, about 30% of the Cl^{36} was unaccounted for. In an attempt to locate the missing Cl^{36} , both the Type 304 stainless steel transfer tube and rubber flask stopper were sectioned and monitored for Cl^{36} activity. This examination revealed that only a minute amount had deposited in the tube but that, apparently, all of the missing Cl^{36} had reacted with the rubber, either as Cl_2^{36} or HCl^{36} . It was interesting to note that some of the Cl^{36} had diffused into the stopper to a depth as great as 1/8 in. In addition, small, highly corroded surface areas existed on the stainless steel transfer tube at each spot that Cl^{36} was found; the relationship between these corroded areas and the deposition, if any, is not known.

Having found the reason for the Cl^{36} inventory shortage, attempts were then made to seal the stopper surface with a heat-resisting, pliable, HCl-resistant material. Several coating materials were tried and found to be unsatisfactory. Finally, the rubber stopper was wrapped in aluminum foil since it was believed that should the Cl^{36} react with the aluminum, it would not diffuse into the metal and a semi-quantitative evaluation of its presence would still be possible. Results of the next MgCl_2^{36} test proved this supposition to be true since 97% of the Cl^{36} was located. It was interesting to note that even after wrapping the stopper a small amount of the Cl^{36} escaped to the rubber through the tubing penetration in the foil and reacted with the surface material. The data for the last three tests are not given in Table 10 since, except for the activity found on the stopper, the test data were similar to the previous data.

6. Discussion of Laboratory Carryover Tests

As a result of these experiments, it was generally concluded that: large amounts of HCl are produced when either MgCl_2 solutions or low-pH solutions of Cl^- are evaporated to dryness; general corrosion and/or pitting of stainless steel surfaces fosters deposition of chlorides; and the exact composition of contaminants must be known when chloride-related stress corrosion is considered, or uncertainty will exist as to the chemical form of the chlorine. The latter is true even when water or steam purity is measured in a few ppb total solids.

Considerable data exist to support the theory that drying of solutions of both HCl and MgCl_2 will produce HCl. Perry³ lists the partial pressure of HCl over a 40% solution of HCl in water as 830 mm of Hg at 35°C. Such values clearly indicate that a droplet of low-pH water containing Cl^- would produce HCl gas, as well as steam, if drying were attempted, unless the Cl^- was precipitated as a salt before a high concentration of HCl occurred. This latter situation would occur if the water contained relatively large amounts of salt-forming cations which form only slightly soluble chloride salts.

The production of HCl by drying a solution of MgCl_2 is considered unavoidable by some chemists. For instance, the comment "The hydrated salt ($\text{MgCl}_2 \cdot n\text{H}_2\text{O}$) cannot be dehydrated without a loss of HCl by simple heating..."⁴ is fairly typical of their opinions. As such hydrates form unavoidably when solutions of Mg^{++} and Cl^- are concentrated, the production of HCl gas from solutions of MgCl_2 also appears to be unavoidable. Therefore, both literature and laboratory data support a theory voiced earlier that large fractions of the Cl^- ³⁶ used in some of the steam loop tests had

acquired the form of HCl^{36} or Cl_2^{36} , as the injected solution of salts was evaporated and then superheated.

L. Chlorine-36 Deposition Test No. 12 - Use of CuCl_2^{36}

1. Test Objectives

This test was designed to determine if any difference exists in the deposition behavior of various chloride salts. The salt used in Test No. 12 was CuCl_2^{36} , added in the form of a dilute solution.

2. Test Conditions

The steam temperatures, pressure, and velocities were typical of several low-velocity, high-temperature tests which have been conducted previously. The loop pressure was approximately 900 psig and the steam temperature (of steam passing over the cartridge heaters) varied from about 537°F (a few degrees of superheat) to a maximum of 900°F at the loop exit. Except for the addition of the three microcuries of Cl^{36} , as CuCl_2^{36} , the steam condition was as-received from the power plant saturated steam drum. No wet steam was allowed to impinge on the heaters during this test.

Prior to insertion in the steam loop, the three 3/4-in.-OD Inconel-clad cartridge heaters were polished with 600-grit emery paper. When placed in the loop, the surfaces of all three heaters were bright, unoxidized, and had almost a mirror-like finish. During this test, the surface temperatures of these heaters ranged from about 700°F to 1100°F . When removed from the loop after the test, the surfaces ranged from a somewhat bright, slightly oxidized condition to light brown and violet in color.

3. Test Procedure

In general, the test procedure duplicated the procedure for Test No. 11 very closely. A pre-test stabilization period was run, the CuCl_2^{36}

solution was injected over a period of several hours, and then a post-test injection system cleaning operation completed the test; the first and last steps involved loop operation with the injection pump adding distilled water to the steam flow. As before, after the cleaning operation was completed, the loop was shut down and the three cartridge heaters were removed for inspection, counting and cleaning.

4. Test Results

The test data were obtained by collecting condensate samples at the loop exit before, during, and after the CuCl_2^{36} addition, by scanning the cartridge heater surfaces after removal from the loop, and by counting all Cl^{36} removable from the cartridge heaters and preheater. As before, steam condensate samples were collected for several days.

A plot of the effluent concentration is shown in Fig. 33. Approximately 30% of the $3.0 \mu\text{c}$ passed through the loop during the actual injection. Another 40% passed through the loop during the injection equipment purging and cleaning process. This was the most rapid throughput for any test where a salt was used. In fact, the effluent concentration curve resembled those obtained when low-pH solutions or HCl^{36} were injected.

A tabulation of the activity levels found on the cartridge heater surfaces is shown in Table 11. It can be seen from this tabulation that the deposition was somewhat uniform except for areas about spacers and thermocouples. No presence of concentrated deposits which could have resulted from wet steam were seen.

A plot of the average circumferential activity found on the cartridge heater surfaces is shown in Fig. 34. The usual small amount of

Table 11 TABULATION OF CARTRIDGE HEATER ACTIVITY LEVELS-TEST NO. 12

Angle from reference, °	Distance from inlet end, in.													
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>	<u>25</u>
<u>Upstream or first heater (downflow)*</u>														
0	138	140	139	114	125	135	178	<u>152</u>	<u>171</u>	<u>185</u>	<u>167</u>	<u>189</u>	208	-
90	128	<u>172</u>	<u>153</u>	<u>161</u>	144	<u>153</u>	<u>240</u>	<u>144</u>	121	<u>129</u>	<u>150</u>	<u>148</u>	201	-
180	<u>148</u>	<u>148</u>	<u>146</u>	<u>122</u>	<u>148</u>	<u>113</u>	<u>144</u>	97	115	114	110	135	212	-
270	<u>133</u>	133	117	90	101	135	169	130	121	135	111	112	<u>227</u>	-
<u>Second heater (upflow)**</u>														
0	-	175	143	116	141	<u>153</u>	<u>126</u>	135	103	119	<u>112</u>	<u>121</u>	94	102
90	-	<u>290</u>	<u>154</u>	<u>149</u>	<u>148</u>	<u>118</u>	<u>108</u>	109	128	<u>182</u>	<u>94</u>	<u>120</u>	103	101
180	-	<u>227</u>	<u>145</u>	<u>142</u>	<u>118</u>	115	110	110	96	<u>101</u>	109	112	<u>111</u>	93
270	-	261	129	127	113	106	119	<u>136</u>	<u>137</u>	126	105	97	<u>98</u>	<u>116</u>
<u>Downstream or third heater (downflow)***</u>														
0	102	105	116	<u>127</u>	110	107	99	99	<u>118</u>	85	105	106	102	-
90	102	94	123	<u>116</u>	117	<u>120</u>	125	103	<u>90</u>	105	98	<u>145</u>	<u>135</u>	-
180	108	<u>130</u>	119	117	<u>178</u>	<u>116</u>	127	<u>123</u>	114	<u>116</u>	106	<u>101</u>	<u>127</u>	-
270	<u>121</u>	<u>103</u>	<u>130</u>	125	<u>126</u>	106	<u>128</u>	<u>99</u>	90	<u>103</u>	<u>107</u>	109	99	-

* Background = 95 counts.

** Background = 101 counts.

*** Background = 96 counts.

contamination was found on all surface areas. As before, areas about spacers and thermocouples collected more Cl^{36} than general surface areas. This increased deposition is exemplified by the peaks shown in Fig. 34. No relationship between deposition and either steam or surface temperatures was noted.

5. Discussion of Test Results

The deposition behavior of the CuCl_2^{36} did not appear to deviate greatly from that of NaCl^{36} . While the injected material passed through the loop more rapidly than did the NaCl^{36} , and the heaters were therefore subjected to a greater throughput of Cl^{36} , the ratio of the amount deposited to the total throughput to which the heaters were subjected appeared to be representative of earlier tests with NaCl^{36} .

It was found that the first heater collected $9.3 \times 10^{-4} \mu\text{c}$, the second collected $5.7 \times 10^{-4} \mu\text{c}$, and the third collected $4.6 \times 10^{-4} \mu\text{c}$; these values were typical of deposition values obtained for NaCl^{36} deposition tests under similar heater surface and test conditions.

During Test No. 12, eight $1 \times 1/2 \times 1/16$ -in. specimens of various materials were suspended in the flowing steam near the loop exit. The steam had been superheated approximately 350°F at this point. After the test had been completed, the specimens were removed and later scanned for Cl^{36} activity by a scintillation detector. No activity was found and no reason for this absence is known unless it can be attributed to the extremely turbulent flow over these local obstructions or to the slight vibration which such specimens are assumed to experience. This absence may explain the general absence of activity on the charged plates which were exposed during earlier tests.

M. Chlorine-36 Deposition Test No. 13 - Use of FeCl_3^{36}

1. Purpose

This test was also designed to determine if any difference exists in the deposition behavior of various chloride salts. The salt used in Test No. 13 was FeCl_3^{36} , which was injected into the loop steam as a dilute, slightly subcooled solution.

2. Test Conditions

The steam temperatures, pressure, and velocities used in this test were typical of several low-velocity, high-temperature tests which had been conducted previously. The loop pressure was approximately 900 psig and the steam temperature (of steam passing over the cartridge heaters) varied from about 537°F (a few degrees of superheat) to a maximum of 900°F at the loop exit. Except for the addition of the three microcuries of Cl^{36} as FeCl_3^{36} , and a few degrees of superheat, the steam conditions were as-received from the power plant saturated steam drum. No wet steam was allowed to impinge on the heaters during this test.

Prior to insertion in the steam loop, the three 3/4-in.-OD Inconel-clad cartridge heaters were polished with 600-grit emery paper. When placed in the loop, the surfaces of all three heaters were unoxidized and had a bright, mirror-like finish. During the test, the surface temperatures of these heaters ranged from about 700°F to 1100°F. When removed from the loop after the test, the heater surfaces ranged from a somewhat bright, slightly oxidized condition, to light brown and violet in color.

3. Test Procedure

In general, the test procedure duplicated the procedure for Test No. 12 very closely. A pre-test stabilization period was run, the

FeCl_3^{36} solution was injected over a period of several hours, and then a post-test injection system cleaning operation completed the test; the first and last steps involved loop operation with the injection pump adding distilled water to the steam flow. As before, after the cleaning operation was completed, the loop was shut down and the three cartridge heaters were removed for inspection, counting and cleaning.

4. Test Results

The test data were obtained by collecting condensate samples at the loop exit before, during, and after the FeCl_3^{36} addition, by scanning the cartridge heater surfaces after removal from the loop, and by counting all Cl^{36} removable from the cartridge heaters and preheater; steam condensate samples were collected for several days.

A plot of the effluent concentration is shown in Fig. 35. Approximately 40% of the $3.0 \mu\text{c}$ passed through the loop during the actual injection and another 30% passed through the loop during the injection equipment purging and cleaning process. This was the most rapid throughput for any test where a salt was used. It was even more rapid than when the CuCl_2^{36} was used. Once again, the effluent concentration curve resembled those obtained when low-pH solutions of chloride salts or HCl^{36} were injected into the loop.

A tabulation of the activity levels found on the cartridge heater surfaces is shown in Table 12. It can be seen from this tabulation that the deposition was somewhat uniform except for areas about spacers and thermocouples. No presence of concentrated deposits which could have resulted from wet steam were seen.

Table 12 TABULATION OF CARTRIDGE HEATER ACTIVITY LEVELS-TEST NO. 13

Angle from reference, °	Distance from inlet end, in.												
	<u>-1</u>	<u>1</u>	<u>3</u>	<u>5</u>	<u>7</u>	<u>9</u>	<u>11</u>	<u>13</u>	<u>15</u>	<u>17</u>	<u>19</u>	<u>21</u>	<u>23</u>
<u>Upstream or first heater (downflow)*</u>													
0	-	105	108	114	114	119	167	<u>132</u>	114	115	<u>142</u>	<u>125</u>	147
90	-	<u>144</u>	<u>127</u>	125	<u>146</u>	<u>127</u>	<u>213</u>	<u>96</u>	101	106	<u>94</u>	<u>98</u>	161
180	-	<u>220</u>	125	<u>135</u>	<u>105</u>	111	<u>132</u>	84	101	<u>126</u>	116	102	<u>247</u>
270	-	<u>128</u>	99	101	115	98	212	98	<u>118</u>	<u>76</u>	100	123	219
<u>Second heater (upflow)*</u>													
0	-	214	118	104	<u>119</u>	112	<u>126</u>	130	113	126	102	105	112
90	-	<u>314</u>	126	<u>125</u>	102	96	<u>106</u>	119	118	100	<u>123</u>	104	98
180	-	<u>239</u>	<u>131</u>	121	98	115	112	<u>155</u>	114	<u>131</u>	122	<u>112</u>	115
270	-	228	125	123	102	<u>122</u>	109	<u>141</u>	<u>120</u>	116	<u>123</u>	99	<u>116</u>
<u>Downstream or third heater (downflow)*</u>													
0	-	<u>131</u>	123	97	104	<u>103</u>	130	96	102	103	<u>124</u>	<u>119</u>	134
90	-	102	117	97	94	<u>94</u>	105	<u>108</u>	111	<u>115</u>	<u>108</u>	89	130
180	-	111	<u>147</u>	98	104	99	125	<u>101</u>	<u>118</u>	<u>103</u>	115	107	146
270	-	106	<u>116</u>	<u>110</u>	<u>120</u>	95	<u>132</u>	111	<u>107</u>	<u>102</u>	93	<u>119</u>	115

* Background = 100 counts.

A plot of the average circumferential activity found on the cartridge heater surfaces is shown in Fig. 36. The usual small amount of contamination was found on all surface areas. As before, areas about spacers and thermocouples collected more Cl^{36} than general surface areas. This increased deposition is exemplified by the peaks shown in Fig. 36. No relationships between deposition and either steam or surface temperatures were noted.

Soaking in 1% HNO_3 and polishing removed 6.2×10^{-4} , 5.3×10^{-4} and 1.2×10^{-4} μc of Cl^{36} from the three cartridge heaters, upstream to downstream, respectively. These amounts were slightly smaller than the 9.3×10^{-4} , 5.7×10^{-4} , and 4.6×10^{-4} μc removed from the heaters after the CuCl_2^{36} test.

5. Discussion of Test Results

The deposition behavior of the FeCl_3^{36} did not appear to deviate greatly from that of NaCl^{36} or CuCl_2^{36} . While the injected material passed through the loop more rapidly than did the NaCl^{36} , and the heaters were therefore subjected to a greater throughput of Cl^{36} , the ratio of the amount deposited to the total throughput to which the heaters were subjected, appears to be representative of earlier tests with NaCl^{36} .

V. DISCUSSION OF TEST RESULTS

Thirteen large-scale steam loop deposition tests and several small-scale bench-type tests were conducted. During these tests, the steam temperature varied between 530 and 900°F, cartridge heater surface temperatures varied between 600 and 1150°F, steam velocities varied between 8 and about 70 ft/sec, and loop pressures varied between 875 and 925 psig. During two tests, wet steam was allowed to impinge on the upstream heater surface. The steam used was typical of conventional power plants except the chloride content was raised slightly by the injection of Cl^{36} and, occasionally, bottled oxygen was added to increase the normal free oxygen content from about 0.005 ppm to about 50 ppm. The heater surface conditions varied from a mirror-like finish (after polishing with 600-grit paper) to a fully-oxidized, pitted condition.

Unfortunately, no method was found to completely prevent the collection of chlorides by nuclear superheater fuel cladding from high purity steam. However, several conditions which definitely increase such deposition were noted. The knowledge of these effects will enable reactor designers to at least alleviate the problems associated with chloride deposition by greatly reducing the rate at which the deposits accumulate. In addition, certain phenomena were noted which may provide a basic step in the understanding of chloride-related stress corrosion.

The sensitivity of the detection techniques enabled detection of as little as 1×10^{-6} gm of Cl^{36} , which was spread over 57 in.² of heater surface area. Occasionally, chlorides were discovered on surfaces which

somewhat bright and apparently free from deposits of any material whatsoever. These data are discussed in detail below. A tabulation of the most significant test data is given in Table 13.

The most significant variable in the deposition behavior described herein was the steam moisture content. The results of allowing wet steam to impinge on heated surfaces is illustrated graphically in Figs. 7 and 25. The greatly increased deposition shown in Fig. 25 resulted from an initial moisture content of 0.4 to 0.5%. Almost at the exact point where sufficient heat had been added to the steam to remove this moisture, the deposition decreased to an almost insignificant amount. A similar effect is shown by the second figure. Here, the bulk steam condition was roughly saturated except for run-down of water droplets from an upstream collection point and the effect of moisture is more pronounced as the water only collected on specific areas of the heater, about spacers and under thermocouple extensions, etc. The circumferential pattern of this deposition can be determined from the data in Table 7 and can be seen on the autoradiograph taken of the deposit as shown in Fig. 22.

It was apparent that steam should be dried thoroughly before being passed over vulnerable components such as superheater fuel cladding. The presence of moisture not only increases the deposition, but it produces a more "permanent" deposit and, presumably, promotes corrosion as well. Certainly, the presence of moisture produces stresses from thermal cycling, as surface areas experience changes from wet to superheated steam flow.

The second most significant factor pertaining to deposition was the surface condition of the collecting component. While rough surfaces would

Table 13 TABULATION OF AMOUNTS OF CHLORINE-36 WHICH DEPOSITED ON THE CARTRIDGE HEATERS

Test No.	Description or Objective of Test	Cl ³⁶ Used	Cl ³⁶ Released (2)	Cl ³⁶ on Preheater	Cl ³⁶ Removed from Upstream Heater, μ c				Cl ³⁶ Removed from Second Heater, μ c				Cl ³⁶ Removed from Downstream Heater, μ c				Collection on Heaters, %		
		μ c	μ c	μ c	H ₂ O Bath	Scrubbing	Acid Soak	Total	H ₂ O Bath	Scrubbing	Acid Soak	Total	H ₂ O Bath	Scrubbing	Acid Soak	Total	Upstream Heater	Second Heater	Downstream Heater
3 ⁽¹⁾	Wet steam at inlet, high velocities	3.0 ⁽³⁾	1.25	-	-	-	0.01400	0.01400	-	-	0.00060	0.00060	-	-	0.00140	0.00140	1.1	0.048	0.11
4	Dry steam at inlet, high velocities	3.0 ⁽⁴⁾	1.95	0.00013	0.00068	-	0.00026	0.00094	-	-	0.00056	0.00056	0.00311	-	0.00033	0.00344 ⁽⁵⁾	0.048	0.029	0.18
5	Dry steam slightly higher temperatures	3.0 ⁽⁴⁾	1.30	0.36	0.00034	0.00153	0.00009	0.00196	0.01360	0.00047	0.00116	0.01523 ⁽⁵⁾	0.00015	0.00037	0.00015	0.00067	0.15	1.20	0.052
6	Repeat No. 4 with different heater finishes	3.0 ⁽⁴⁾	1.10	-	-	0.00047	0.00043	0.00090 ⁽⁷⁾	-	0.00040	0.00036	0.00076	-	0.00397	0.02270	0.02667 ^(5,6)	0.093	0.069	2.8
7	High-temperature test, low velocity steam	3.0 ⁽⁴⁾	0.18	-	-	0.00014	0.00013	0.00027	-	0.00026	0.00012	0.00038	-	0.00013	0.00007	0.00020	0.15	0.21	0.11
8	Repeat of No. 7, new Inconel heaters	3.0 ⁽⁴⁾	1.31	-	-	-	-	0.00072	-	-	-	0.00043	-	-	-	0.00114	0.055	0.033	0.087
9	Exposed numerous surface conditions for autoradiographic purposes	3.0 ⁽⁴⁾	-(8)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
10	MgCl ₂ ³⁶ was used	3.0 ⁽⁴⁾	2.6 ⁽⁹⁾	-	-	-	0.00033	0.00033	-	-	0.00009	0.00009	-	-	0.00012	0.00012	0.013	0.0035	0.0046
11	The effect of solution pH was investigated	3.0 ⁽⁴⁾	-	-	-	-	0.00014	0.00014	-	-	0.00016	0.00016	-	-	0.00003	0.00003	-	-	-
12	CuCl ₂ ³⁶ was used	3.0 ⁽⁴⁾	2.1 ⁽⁹⁾	-	-	-	0.00093	0.00093	-	-	0.00057	0.00057	-	-	0.00046	0.00046	0.044	0.027	0.022
13	FeCl ₃ ³⁶ was used	3.0 ⁽⁴⁾	2.1 ⁽⁹⁾	-	-	-	0.00062	0.00062	-	-	0.00053	0.00053	-	-	0.00012	0.00012	0.030	0.025	0.0057

(1) Tests No. 1 and No. 2 were exploratory and are not reported.

(2) This amount refers to the amount which was released while the cartridge heaters were in the loop and approximately the amount which flowed over each heater.

(3) Over a period of three days.

(4) Injection completed in one day.

(5) Same heater, pitted surface.

(6) Approximately 20% of the activity remained after the cleaning procedures.

(7) Polishing removed 0.000132 μ c.

(8) No quantitative data were sought during this test.

(9) The throughput was believed to have been affected by a preheater change.

be expected to collect more impurities than smooth surfaces, an added factor appears to exist. This factor is concerned with the method by which the roughness had been produced. For instance, mechanically produced surface defects collected much less Cl^{36} activity than did small corrosion pits and blemishes which had been produced by chemical attack. Autoradiographs clearly indicated that defects from either chemical or corrosion attack collected easily detectable deposits of Cl^{36} while purely mechanically produced defects did not collect sufficient Cl^{36} deposits to enable clear-cut detection.

The relative significance of specific surface defects and physical barriers was exemplified by the results for Test No. 9 (see Fig. 25). During this test, a heavily oxidized heater with nine attached rings collected only a small fraction of the Cl^{36} collected by a pitted heater. While all spacers and thermocouples increased the local deposition of Cl^{36} , it is not known whether this increase was due to physical disturbance of the flow by the protuberance or to a surface change which resulted from the use of a brazing flux, etc.

The data generally indicated that some attracting force existed between the Cl^{36} compounds (or particles) and specific areas on the heater jacket surfaces, particularly the chemically attacked (but not roughened) and pitted areas. If this force was electrostatic, results of tests using charged plates did not substantiate this theory. However, as explained earlier, the absence of any deposition on the charged plates does not preclude such an effect.

The observed accumulation of chlorides in or about pits may explain reports of a relationship between corrosion pits and initiation of stress cracking.^{2,5}

There did not appear to be any correlation between Cl^{36} collection and basic materials. No obvious difference was seen between Types 304 and 347 stainless steels, Inconel, and several low-alloy steel materials. However, the low-alloy steel materials were exposed as small coupons, along with other materials, and none of the coupons exhibited detectable activity after the test. Both types of stainless steel and Inconel were used as heater jacket material.

No effect on Cl^{36} deposition, of either steam or surface temperatures, was noted. However, since temperatures will affect the corrosion rate of superheater materials, the result of operation at different temperatures will surely appear with extended operation.

Increased Cl^{36} deposition effect was noted at low steam velocities. However, reactor coolant velocities will be determined by requirements other than chloride deposition, and since the higher, more realistic velocities give less deposition, the exact relationship between deposition and velocities was given only cursory attention.

In almost all instances, Cl^{36} deposition peaked about spacers and other surface discontinuities. The increase in deposition is generally attributed to local velocity effects around the spacers. Increased Cl^{36} deposition also resulted on surfaces subjected to chemical or corrosion attack. During fabrication of superheater fuel, the clad surface should be cleaned by mechanical means and not by chemical means. Because of the

possible increased deposition of chlorides in chemically treated surfaces, "pickling" of stainless steel piping should be avoided if the piping is to be used under conditions which are conducive to stress chloride corrosion.

No large effect of prior oxidization of surfaces was noted, except where the corrosion had progressed to pitting. It was definitely established that corrosion pits had an affinity for chlorides.

Some differences in the deposition behavior of various chloride compounds were noted during the tests. Injection of solutions of NaCl^{36} (pH~8) resulted in the heaviest deposits; deposition from CuCl_2^{36} and FeCl_3^{36} solutions (in steam) were similar and slower than from the NaCl^{36} ; and the use of either MgCl_2^{36} or low-pH solutions of salts resulted in the least deposition. This general indication can be seen if the data in Table 10 are examined. However, very little significance was attached to this general behavior since: some deposition occurred under all conditions; the paucity of data questions the reliability of these indications; and it may be impossible to predict the molecular form which chlorides will assume when high purity water is dried under high-temperature, high-pressure conditions.

The behavior of MgCl_2^{36} solutions during drying is worth noting. It is believed that this process produced HCl^{36} , which resulted in very little deposition, and that MgCl_2^{36} cannot exist in superheated steam; only MgO and HCl^{36} can exist. If this phenomenon can be proven, then it may be desirable to increase the Mg^{++} concentration of water for the purpose of preventing the formation of chloride salts, by the formation of gaseous HCl . A few parts per billion of HCl in either steam or water would

probably not be undesirable. Certainly, the industry should be aware of the relationship between MgCl_2 in boiler water and the possible production of HCl .

An extremely important phenomenon observed during each test was the magnitude of chloride concentrations during loop transients, as compared to the steady-state values. The effluent concentration peak seen after each loop startup portrays such behavior very clearly. Even after a shutdown of only 30 minutes and only slight cooling of the loop piping, it was impossible to re-start the loop without causing very large concentration changes. Occasionally, the loop effluent Cl^{36} concentration increased to a value which was 100 times larger than the value prior to shutdown. This behavior was typical of the effect of operational changes on the properties of high purity systems and certainly complicates the methods whereby impurities, such as chlorides, are deposited throughout fluid systems. This evidence indicates strongly that:

- 1) Determinations of impurities in reactor systems cannot be divorced from an operating history of the respective system.
- 2) Where the problem of chlorides is of concern, a simple reduction in the normal fluid impurity concentrations may not be greatly beneficial, as the "noise"* levels may negate the effect of these reductions. Indeed, it appears reasonable to expect that a major portion of chloride deposition in nuclear superheaters could result from operational transients, such as startup, and hence

* Changes in impurity concentration due to hideout impurities which are released during operational transients.

be a function of the system geometry, etc., as well as the normal fluid impurity concentration.

This phenomenon was clearly shown in a report which discussed water impurities versus purification system operation in the PWR⁶. For instance, the primary coolant system impurity concentration due to "noise" variations almost completely hid the effect of stopping purification for over 1000 hr.

It is presently believed that minor temperature and pressure changes, power plant load changes, etc., produced large Cl^{36} concentration changes in the loop effluent. Such fluctuations can be seen in Figs. 10 and 14.

VI. CHLORINE-36 INVENTORY RECORD, CONTROL AND DISPOSAL

Because the radioactive Cl^{36} was being used in an unrestricted area, a complete and careful inventory record was kept.

Purchasing, storage, handling, and disposal of the Cl^{36} were accomplished in accordance with the General Licensing Provisions in Title 10 of the Code of Federal Regulations. No more than 10.0 μc of Cl^{36} were on hand at any one time. Care was taken to control all materials being used and prevent contamination of equipment or buildings; a record was kept of use and disposal of the Cl^{36} . No accidents occurred whereby contamination was known to have been spread. After the tests had been completed, all waste, laboratory glassware, rags, etc., were shipped to ORNL for disposal. The loop piping was thoroughly decontaminated with wet, 530°F, 900 psig steam; disposal of the loop will not be accomplished until future non-radioactive tests have been completed. At that time, the internal surfaces will be monitored to determine if special disposal procedures are required.

Forty-three microcuries of HCl^{36} were purchased from ORNL and used during the tests. A bar-graph type inventory record of these 43 μc of HCl^{36} is shown in Fig. 38. It can be seen from this graph that, after the initial purchase of 10.0 μc , allowance was made for Cl^{36} temporarily retained in the test loop, etc., before additional Cl^{36} was purchased.

A block diagram which covers the storage, use, and disposal of the Cl^{36} is shown in Fig. 39. A more detailed description is given in the preceding sections of this report.

Twice during the test program and after completion of all tests, a Radiological Health Consultant from the Pinellas County Health Department

visited the test site and made smear tests. Smears were taken from the floor beneath the loop, in the sample processing area, and from the external surfaces of some test equipment. The total activity of such smears indicated activity levels between 1 and 17 $\mu\text{c}/30$ to 40 in.^2 of surface area. These values were representative of the natural background found in other areas of the plant and, therefore, indicated little or no Cl^{36} contamination.

Even though it was not believed necessary, film badge service was provided for persons involved in the tests. No indication of any dose was reported, except for one period when the reported dosages were between 74 and 100 mrem for one month. This report was considered to be in error since the film badges of several persons who had not been in any radioactive areas also showed the same dosage.

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ILLUSTRATIONS

Fig. 1 Chloride deposition test equipment layout.

Fig. 2 Sketch of cartridge heater which simulates BONUS superheater fuel rod.

Fig. 3 Detector shield assembly and geometry reference source.

Fig. 4 Over-all view of counting equipment.

Fig. 5 Absorption effect of thin plastic on 0.76-Mev beta particles.

Fig. 6 Absorption effect of thin plastic on 0.71-Mev beta particles.

Fig. 7 Longitudinal plot of heater activity (Test No. 3).

Fig. 8 Loop effluent (condensate) activity during and after Test No. 4.

Fig. 9 Longitudinal plot of heater activity (Test No. 4).

Fig. 10 Loop effluent concentration during Test No. 5.

Fig. 11 Comparison of cartridge heater activity after Tests No. 4 and No. 5.

Fig. 12 Plot of cartridge heater activity after Test No. 5.

Fig. 13 Photograph of a badly pitted area which contained a heavy deposit of Cl^{36} . (7X)

Fig. 14 Loop effluent concentration during Test No. 6.

Fig. 15 Plot of cartridge heater activity after Test No. 6.

Fig. 16 Loop effluent concentration during Test No. 7.

Fig. 17 Plot of cartridge heater activity after Test No. 7.

Fig. 18 Loop effluent concentration during Test No. 8.

Fig. 19 Plot of cartridge heater activity after Test No. 8.

Fig. 20 Two-plate electrode assembly used during Test No. 9.

Fig. 21 Loop effluent activity during Test No. 9.

Spot Contour
a. Autoradiograph of wetted area. (1X)

Thermocouple
Spot contour
Downflow →
b. Photograph of wetted area. (about 1.3X)

Fig. 22 Autoradiograph and photograph of section of heater which experienced wet steam flow.

a. Autoradiograph of marred area. (1X)

b. Photograph of marred area. (about 1.3X)

Fig. 23 Autoradiograph and photograph of intentionally marred heater surface area.

a. Partially wetted area (inlet).

b. Area cooled by superheated steam (outlet).

Fig. 24 Autoradiographs of heater surfaces after exposure to steam contaminated with NaCl³⁶.

Fig. 25 Plot of cartridge heater activity after Test No. 9.

Fig. 26 Comparison of same cartridge heater activity levels after two similar tests.

Fig. 27 Loop effluent activity during Test No. 10.

Fig. 28 Plot of cartridge heater activity after Test No. 10.

Fig. 29 Plot of loop effluent activity during Test No. 11.

Fig. 30 Plot of cartridge heater activity after Test No. 11.

Fig. 31 Initial laboratory Cl^{36} release-by-boiling apparatus.

Fig. 32 Laboratory Cl^{36} release-by-boiling apparatus with steam purging provisions.

Fig. 33 Loop effluent activity during Test No. 12.

Fig. 34 Plot of cartridge heater activity after Test No. 12.

Fig. 35 Loop effluent concentration during Test No. 13.

Fig. 36 Plot of cartridge heater activity after Test No. 13.

Fig. 37 Enlarged closeup photograph of typical heater spacer after cleaning.

Fig. 38a Inventory record of Cl^{36} .

Fig. 38b Inventory record of Cl^{36} . (Continued)

Fig. 38c Inventory record of Cl^{36} . (Concluded)

Fig. 39 Diagrammatic description of handling and disposal of Cl^{36} .