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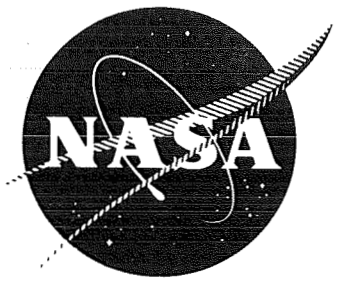

EVALUATION OF TANTALUM FOR MERCURY CONTAINMENT IN THE SNAP-8 BOILER

By

H. Derow, B. E. Farwell, E. B. Johnson, L. A. Kimura
J. C. Whipple, M. K. Wong, D. Yee

AEROJET-GENERAL CORPORATION
Azusa, California

prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

NASA Lewis Research Center

Contract NAS 5-417

Martin J. Saari, Program Manager

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TOPICAL REPORT

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NASA Lewis Research Center
Cleveland, Ohio
Martin J. Saari, Program Manager
SNAP-8 Program Office

FOREWORD

The work described in this report was performed at Aerojet-General Corporation, Nuclear Division, San Ramon, California as part of the SNAP-8 Electrical System Contract being conducted within the Power Systems Department of Aerojet-General Corporation, Azusa, California. The studies and the results pertaining to the evaluation of tantalum as a mercury, containment material in the SNAP-8 boiler is covered in this report. The work was performed under NASA Contract NAS 5-417 with Mr. Martin J. Saari as NASA Program Manager, and Dr. W. F. Banks as Aerojet-General Corporation Program Manager. Aerojet-General Corporation acknowledges P. L. Stone and E. R. Furman of the NASA, Lewis Research Center, for their contributions during performance of the work.

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ABSTRACT

A series of tests were conducted to evaluate tantalum as a material for mercury containment in the SNAP-8 boiler. Mercury corrosion of tantalum was known to be minimal; therefore, the tests were designed to investigate various mechanical designs of the boiler structure containing tantalum. The results of the tests provided information for evaluating the effects on tantalum of air and polyphenyl ether (Mix-4P3E) contaminants in the mercury loop. The test results and their application to the SNAP-8 boiler are described.

SUMMARY

A test program was conducted to evaluate the use of tantalum for mercury containment in the SNAP-8 boiler. Five tests on subscale SNAP-8 simulated boiler tube configurations were conducted. Various candidate basic boiler tube design concepts and fabrication procedures were evaluated. The effects of potential SNAP-8 mercury loop contaminants on tantalum were also evaluated.

Two Hg containment tube configurations were found capable of providing equivalent acceptable reliability for up to 40,000 hours of service in the SNAP-8 boiler. These are, first, a bimetal tube of tantalum bonded by hot coextrusion to the I.D. surface of a 316 stainless steel tube. Second, unbonded non-concentric tubes of tantalum inside a 321 stainless steel tube. In the latter instance the two tubes are thermally bonded through a non-flowing NaK inventory which fills the annular space between the two tubes. In both cases the heat is transferred from a flowing NaK stream on the outside of the stainless steel tube to a flowing mercury stream on the inside of the tantalum tube.

It was established that mercury loop contamination by air or Mix-4P3E, an organic fluid used for lubrication and cooling of various components in the SNAP-8 system, can detrimentally affect the heat transfer capability of a SNAP-8 boiler. The reduced heat transfer is caused by a thermal barrier formed on the tantalum surface by a reaction between the tantalum and the contaminant. This barrier can be effectively removed by chemical cleaning.

I. INTRODUCTION

SNAP-8 is a 35 kw nuclear-electric power conversion system for use in space. The system operates on a mercury (Hg) Rankine cycle using NaK (eutectic sodium -- potassium mixture) as the heat-input and heat-rejection working fluid. The power conversion system is being developed by Aerojet-General Corporation (AGC) for the National Aeronautics and Space Administration. The nuclear reactor is being developed for the Atomic Energy Commission by Atomics International.

The power conversion system, Figure 1, uses mercury as the working fluid and is coupled to the reactor cooling loop by a heat exchanger (boiler) where the mercury is preheated, vaporized, and superheated. The superheated vapor drives a turbine-alternator assembly which develops 35 kw of useful electrical power. The saturated mercury vapor leaving the turbine passes through a condenser and then to a mercury pump to complete its cycle. Cooling for the condenser is provided by a pump-driven NaK loop which couples the condenser and space radiator. Lubrication and cooling of the system is provided by a loop using a polyphenyl ether (Mix-4P3E) organic working fluid. Lubrication is provided for the bearings in the turbine-alternator assembly and the mercury pump-motor assembly. Cooling is provided for the alternator, pumps, and electrical controls. The Mix-4P3E loop is pump driven and has its own heat-rejection radiator.

The boiler for the SNAP-8 system was for several years constructed of 9 Cr-1 Mo steel. After a considerable number of capsule corrosion tests and forced-flow subscale and full scale tests had been conducted, it was apparent that this material was not sufficiently resistant to mercury corrosion at 1100°F, the maximum liquid mercury temperature for SNAP-8 boiler operation, to provide the required minimum 10,000-hour life. A reliable approximation of the mercury corrosion resistance of a given material can be made by examining the solubility in mercury of the major alloy elements in the material. Figure 2 shows the solubility of several elements in liquid mercury as a function of temperature. It can be seen that although the solubility of iron is low, the refractory metals are only slightly soluble in mercury, with the

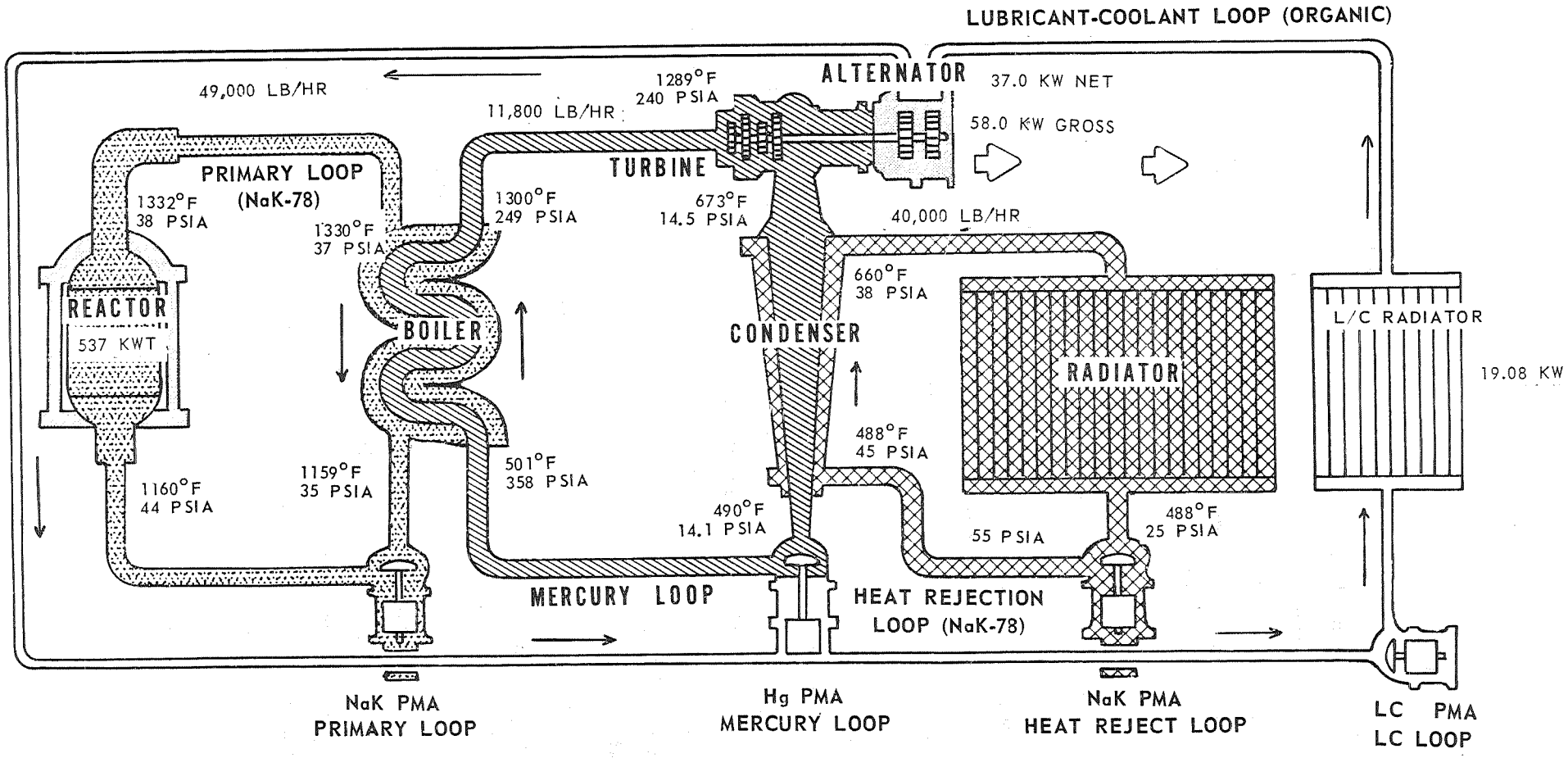


Figure 1. SNAP-8 System Schematic

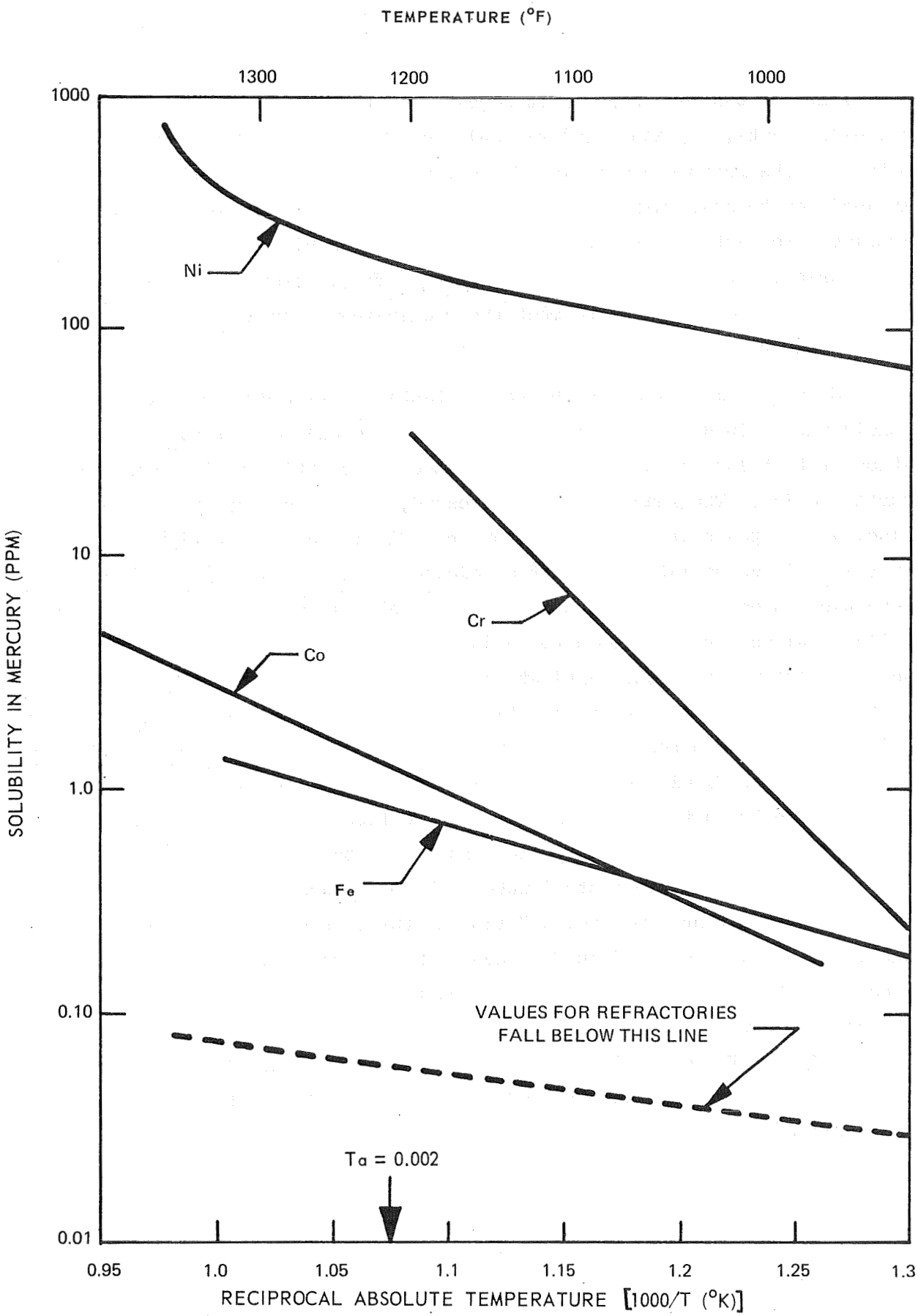
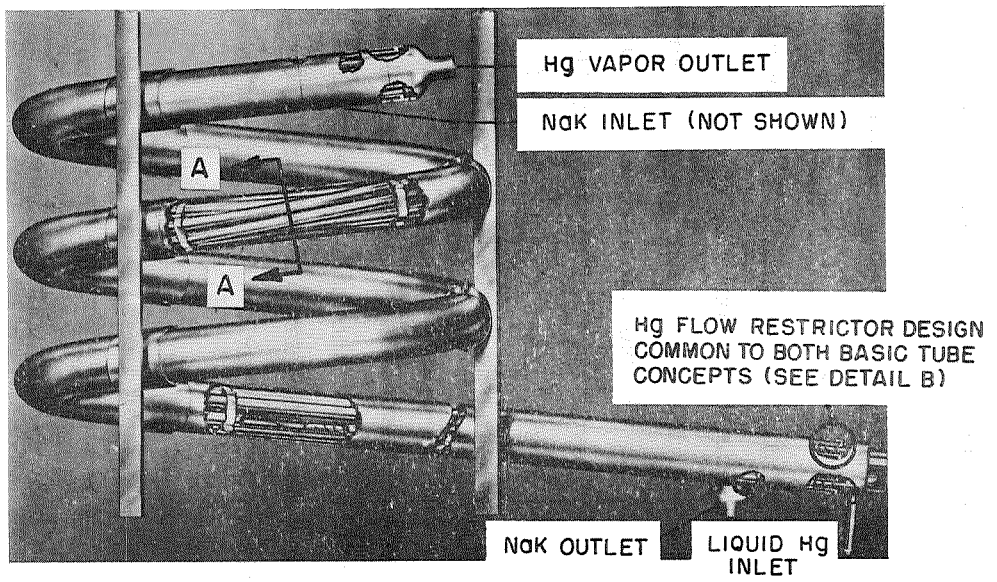


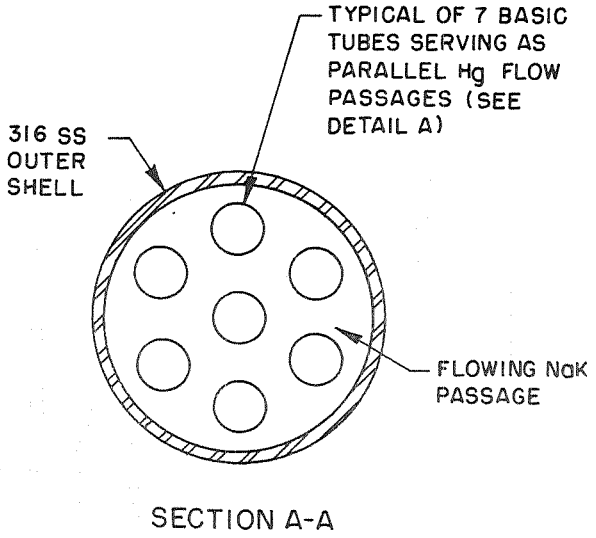
Figure 2. Solubility of Some Elements in Mercury

solubility of tantalum being just barely detectable. Based primarily on this solubility data, tantalum (Ta) was selected from the available refractory metals as the reference material for mercury containment in the SNAP-8 boiler. In addition to exhibiting the lowest solubility in mercury of the commercially available refractory metals, tantalum has a proven history of producibility and fabricability in the chemical industry. The mechanical properties, though data was limited, appeared to meet the requirements for a 10,000 hour life SNAP-8 boiler.

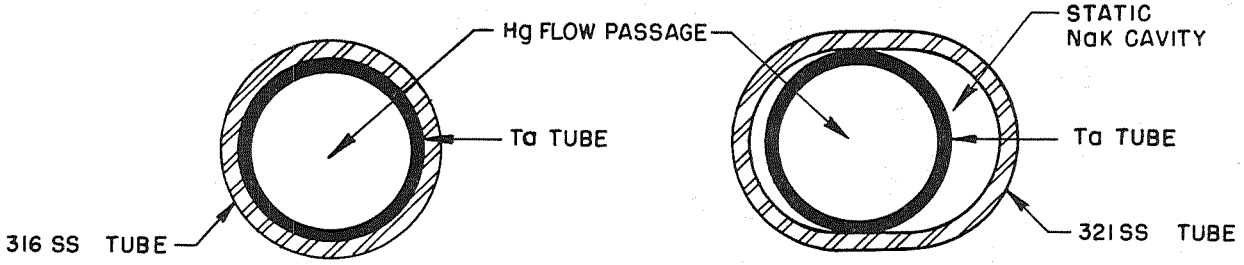
Several limitations in the use of tantalum existed which required special consideration in designing the material into the SNAP-8 system. Air contamination of tantalum exposed at 400°F resulted in formation of a non-adherent surface oxide. The reaction rate increased, with increased exposure temperature. Also, it was postulated that tantalum in a flowing SNAP-8 multi-metal NaK system would accumulate interstitial elements, carbon and oxygen, from the NaK resulting in embrittlement and accelerated metal mass transfer. Two basic boiler designs were developed to avoid the above uncertainties. In both designs the tantalum was protected against elevated temperature air oxidation, by fabricating oxidation resistant 316 stainless steel (316 SS), all sections of the boiler which were exposed to the air. Also the tantalum was protected against exposure to the flowing NaK by isolation utilizing a stainless steel tube positioned between the tantalum tube and the flowing NaK side of the boiler. One design provided for mercury flow inside the tantalum tube and NaK flow on the outside of a stainless steel tube. The other design provided for mercury flow inside the tantalum tube, NaK flow on the outside of the stainless steel tube, and a static NaK annulus between them. Figure 3 depicts the two designs. Since both boiler designs were considered to be of equal potential, it was decided to test in a subscale SNAP-8 configuration both mercury containment tube designs. The objectives of the test program were as follows:

- Evaluate the heat transfer performance of two alternate mercury containment tube designs.
- Evaluate the effect of SNAP-8 operating conditions on the structural integrity of the mercury containment tube designs.





SECTION A-A



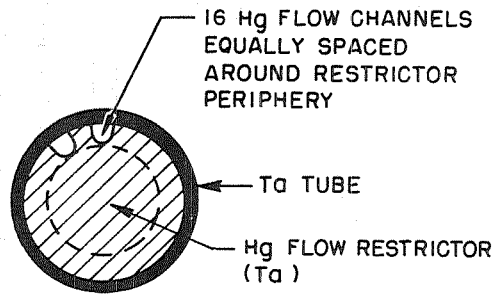
BASIC CONCEPT NO. 1

CONCENTRIC Ta 316-SS TUBES IN INTIMATE CONTACT

BASIC CONCEPT NO. 2

NON-CONCENTRIC Ta AND 321 SS TUBES CONTAINING STATIC NaK ANNULUS

DETAIL A



DETAIL B

Figure 3. SNAP-8 Boiler Showing Alternate Concepts of Hg Containment Tube Design

. Establish the mercury-side corrosion effects on the materials.

. Evaluate the effect on tantalum tube heat transfer performance, of the presence of air or Mix-4P3E in the mercury system.

. Select the optimum design for full scale SNAP-8 boiler use.

II. TEST SECTION DESCRIPTION

The SNAP-8 boiler is a counterflow tube-in-tube heat exchanger. The mercury flows inside seven parallel tubes which are interconnected by manifolds at the mercury inlet and outlet of the boiler. The seven tubes are bundled inside a NaK containment shell. The NaK flows through the annular space formed between the tube bundle and the outer shell (see Figure 3). Approximately the first 4 feet of the SNAP-8 boiler length is considered the most critical for achieving adequate boiler performance. Within this length, the major portion of total heat transfer from the flowing NaK to flowing mercury must occur to meet the desired design criteria of total hydraulic pressure drop through the boiler, and to insure minimum liquid mercury carry-over to the turbine. This section of the tantalum tube contains a solid tantalum bar with channels machined into the outside surface. The only mercury flow passage through the tube are the channels in the bar by virtue of a no-gap fit between the tube I.D. and bar O.D., achieved by cold swaging of the tube over the bar. The liquid mercury reaches its highest temperature of approximately 1100°F in this initial tube length in the process of being converted to vapor. Therefore, any acceptable basic tube design for the boiler must be capable of effectively transmitting the heat through its wall and it must withstand the corrosion and erosion potential of the high temperature mercury flow. For this reason, the test program concentrated primarily on evaluation of test sections simulating this initial four foot section of the overall boiler length.

Two designs of the mercury containment tube were evaluated (see Figure 3). Tests were performed on both designs using single mercury containment tube test sections to simplify fabrication. One tube configuration consisted of concentric 316 stainless steel (outside) and tantalum tubes (Basic Concept No. 1 in Figure 3). The other tube configuration consisted of non-concentric

321 stainless steel (outside) and tantalum tubes with a static NaK inventory in the annular space between them (Basic Concept No. 2 in Figure 3). Basic Concept No. 1 which was used for construction of four of the five test sections evaluated were fabricated by cold swaging, hot co-extruding, and explosively bonding the tubes (see Table I). The last test section, representing Basic Concept No. 2, was assembled as shown in figure 3 with the tantalum and stainless steel tubes providing the necessary stand-off distance for the static NaK containment.

A. UNBONDED CONCENTRIC TUBES

Test Sections Numbers 1 and 2 (see Figure 4) contained Hg containment tubes of bimetal tube construction. The bimetal tube consisted of concentric tantalum and 316 stainless steel tubes in intimate contact, but not metallurgically bonded. The intimate contact of the bimetal tube was produced by cold swaging. To prevent air exposure of the Ta during system operation, and the resultant catastrophic oxidation, the tantalum tube was located entirely within the 316 stainless steel envelope. The load-carrying structure of the section, and of a full size SNAP-8 boiler containing this tube concept, was of welded 316 stainless steel. The two test sections, Numbers 1 and 2, were identical except that the latter contained a mechanical seal at the mercury inlet end to prevent mercury flow through a gap between the tantalum and 316 tubes which formed when the section had reached operating temperature. This gap results from the significantly different thermal expansion coefficients of the two materials between 75° and $1300^{\circ}F$, 3.8×10^{-6} and 10.3×10^{-6} in./in./$^{\circ}F$, respectively. Free movement of each relative to the other allowed the 316 stainless steel to pull away from the tantalum forming a 0.0015 inch radial gap at $1100^{\circ}F$, the boiling temperature of the mercury in the test section. This free relative movement also avoided the necessity of compensating for high mechanical loads which might have been applied had the tantalum and 316 stainless steel been structurally connected.

B. BONDED CONCENTRIC TUBES

Test Section Numbers 3 and 4 (see Figure 5), contained concentric tantalum and 316 stainless steel tubes which were metallurgically bonded into

TABLE I

TEST SECTION DESIGN SUMMARY

Test Section Number	1	2	3	4	5
AGC Designation	4C-1	4C-1A	4C-8	4C-9	SB-1
Basic Tube Design Concept(1)	1	1	1	1	2
Mercury Tube Type(1)	Unbonded Ta/316 SS	Unbonded Ta/316 SS	Bonded(5) Ta/316 SS	Bonded(5) Ta/316 SS	Unimetal Ta
Ta/316 SS Tube Fabrication Procedure	Cold Swaged(2)	Cold Swaged(2)	Hot-Co-Extruded	Explosively Bonded	(3)
Mercury Flow Restrictor Material	Tantalum	Tantalum	Tantalum	Tantalum	Tantalum
Test Section Length (in.)	50	50	44	44	360
SNAP-8 Scale Factor (Based on Total SNAP-8 Boiler Hg Flow)	1/22	1/22	1/22	1/22	1/7(4)

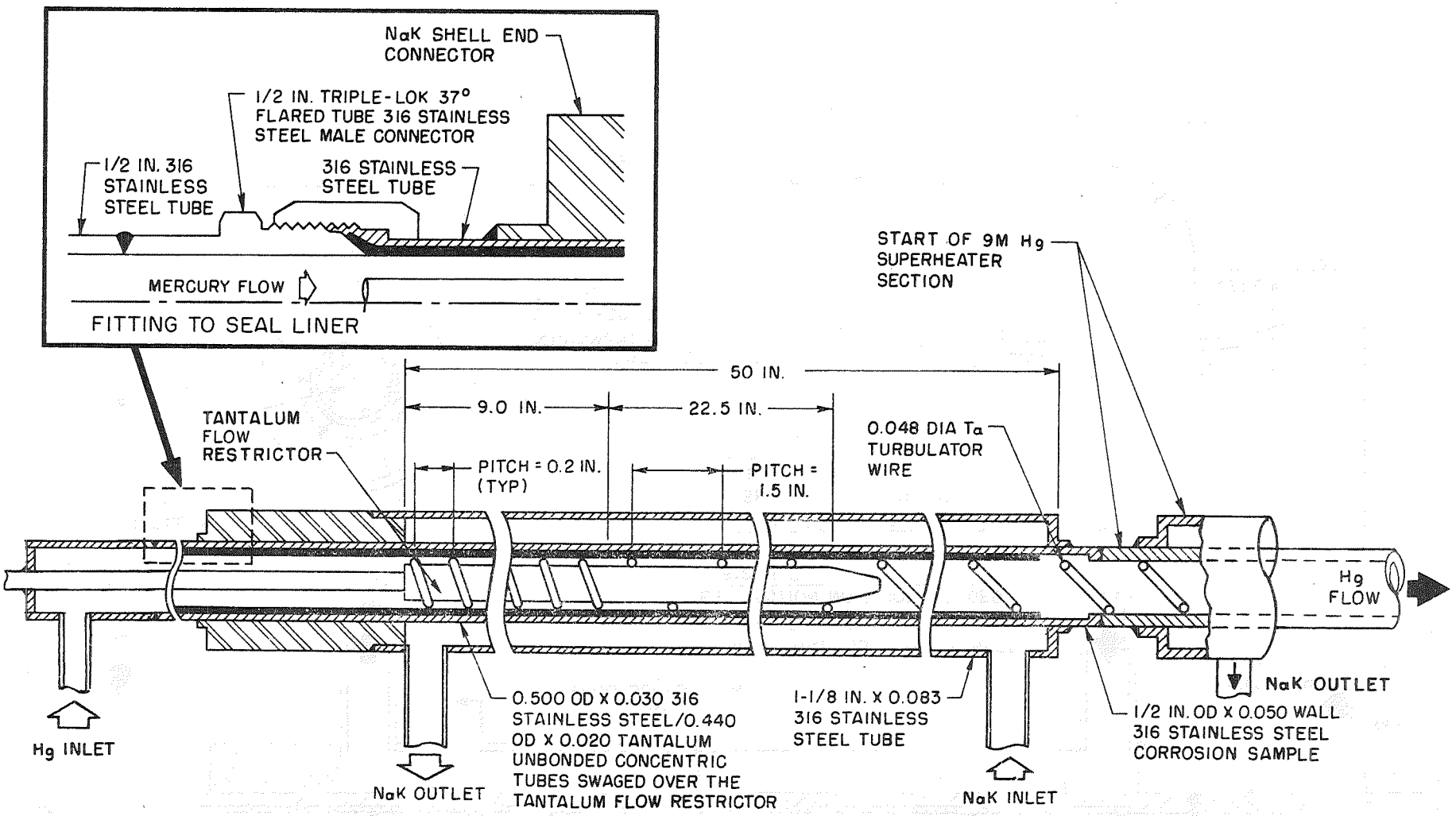
(1) See Figure 3 for pictorial description.

(2) These sections contained tantalum plugs which formed the mercury flow channel by helically winding a wire on the plug O.D. rather than machining grooves to the O.D. surface as is shown in Figure 4.

(3) Stainless steel and tantalum tubes were assembled to provide the necessary stand-off distance for static NaK containment.

(4) This test section represented a full size replica of one of the seven SNAP-8 boiler mercury containment tubes.

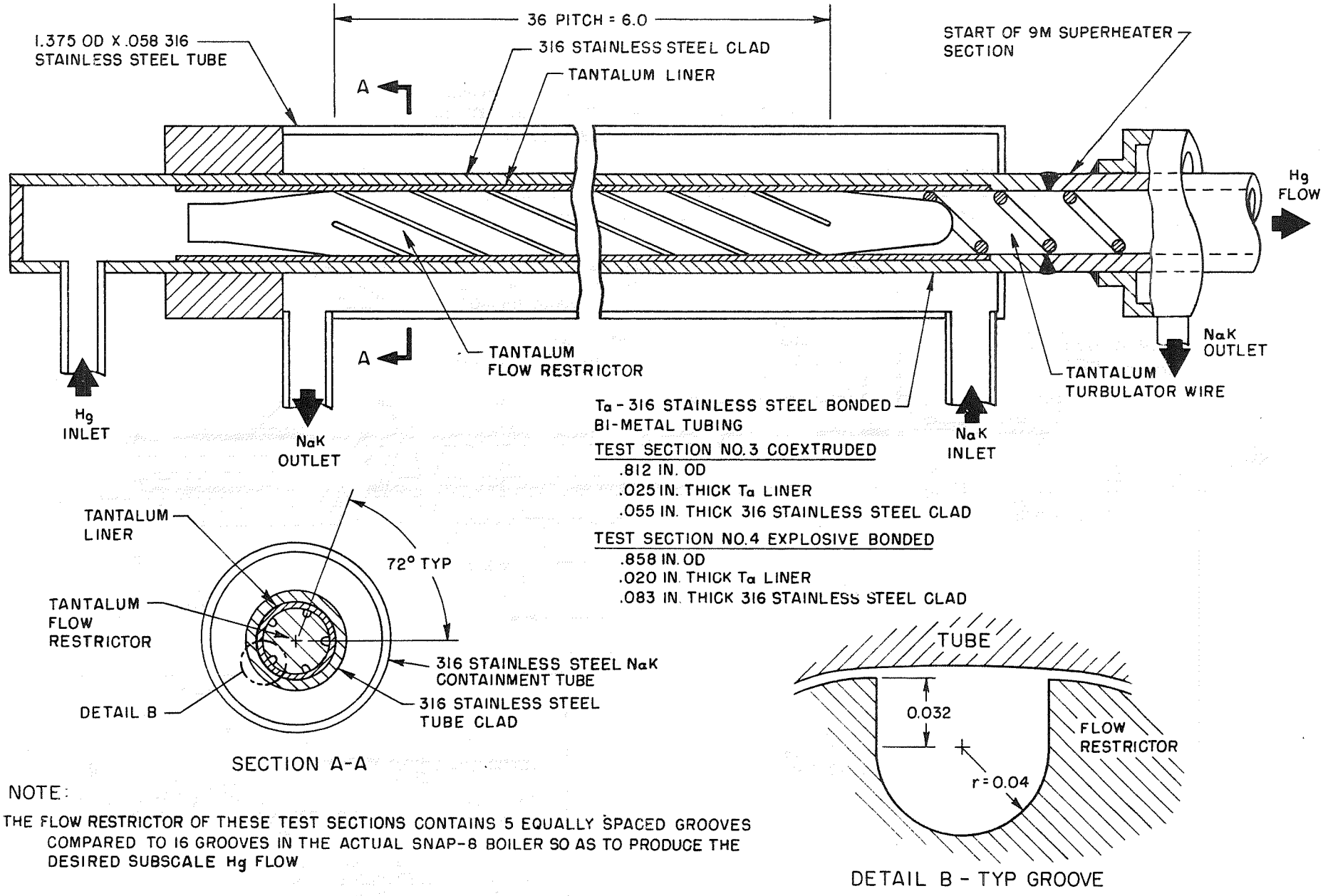
(5) Metallurgically bonded.



NOTES: (1) BASIC DESIGN OF TEST SECTIONS NO. 1 AND 2 ARE IDENTICAL EXCEPT FOR FEATURE SHOWN IN INSERT. TEST SECTION NO. 2 CONTAINED THE DESCRIBED MECHANICAL SEAL TO PREVENT MERCURY FLOW THROUGH THE GAP BETWEEN THE TANTALUM AND 316 STAINLESS STEEL EXISTING AT OPERATING TEMPERATURE OF APPROXIMATELY 1100°F.

(2) THE TANTALUM TURBULATOR WIRE CAUSES VORTEX FLOW OF THE LIQUID MERCURY REQUIRED TO MAINTAIN CONTACT BETWEEN THE MERCURY AND TANTALUM TUBE WALL. THIS FLOW ALONG THE WALL FACILITATES MAXIMUM HEAT TRANSFER FROM THE NaK THROUGH THE TUBE WALL TO THE MERCURY.

Figure 4. Unbonded Cold-Swaged Test Sections 1 and 2 Bimetal Ta-316 Stainless Steel Hg Containment Tube



NOTE:

THE FLOW RESTRICTOR OF THESE TEST SECTIONS CONTAINS 5 EQUALLY SPACED GROOVES COMPARED TO 16 GROOVES IN THE ACTUAL SNAP-8 BOILER SO AS TO PRODUCE THE DESIRED SUBSCALE Hg FLOW

Figure 5. Test Sections No. 3 and 4 - Bonded Bimetal Ta-316 Stainless Steel Tube

an integral bimetal tube construction to prevent separation during boiler start-up and/or continuous heating at the operating temperature. The bonded tube of Test Section No. 3 was produced by hot coextrusion by Nuclear Metals Division, Whittaker Corp., Concord, Mass. The bonded tube of Test Section No. 4 was produced by explosive bonding by Aerojet-General Corporation, Downey, California. Catastrophic air exposure of the tantalum was avoided by the design utilized in Test Section No. 1. However, the bond between the tantalum and 316 stainless steel tubes was sufficiently strong to avoid the radial gap. The tantalum tube yielded so as to expand with the 316 stainless steel during heating. The basic structure of a full size SNAP-8 boiler containing this tube concept would be 316 stainless steel with the tantalum removed on the tube ends, thus avoiding the problem of joining the tantalum to the 316 stainless steel. As was true of the radial expansion, the bond was sufficiently strong to avoid failure due to detrimental longitudinal loads.

C. NONCONCENTRIC TUBES WITH STATIC NaK CAVITY

The mercury containment tube of Test Section No. 5 (see Figure 6) was made of Ta and the flowing NaK outer containment shell was made of 316 SS. The Ta was protected from the flowing NaK by a 321 SS tube which enclosed the Ta tube forming an annular cavity filled with static NaK. In a full size SNAP-8 boiler containing this design concept the entire mercury containment system, including mercury inlet and outlet manifolds would be constructed of tantalum. To avoid continuation of tantalum outside of the boiler envelope formed by the 316 stainless steel outer shell the Ta is joined to 316 stainless steel piping, providing mercury containment outside the boiler, by means of a tantalum/316 stainless steel transition joint (see Figure 6). The 316 stainless steel outer shell and 321 stainless steel flattened-oval tube are welded to the 316 stainless steel side of the transition joint. Thus, the tantalum structure is free to move independently of the combined 316/321 stainless steel structure of the boiler but the Ta and stainless steel structures are tied together through the two tantalum/316 stainless steel transition joints. The 321 SS tube is of flattened-oval cross-section to accommodate the relative diametral change between the Ta and stainless steel portions of the SNAP-8

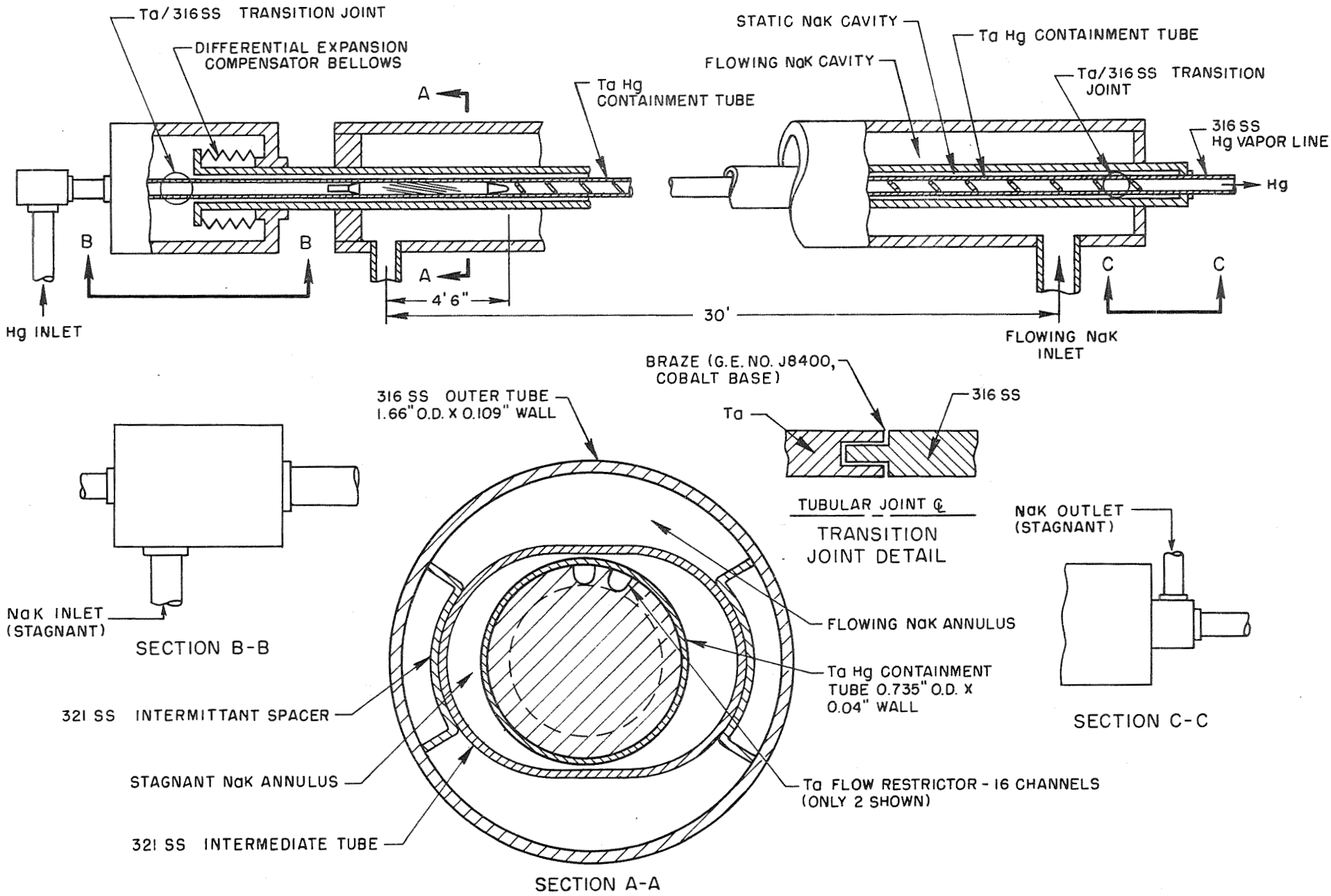


Figure 6. Test Section No. 5 - Unimetal Tantalum Mercury Containment Tube with 321 Stainless Steel Tube Forming Static NaK Cavity

coiled boiler configuration due to differential thermal expansion as the boiler heated up to operating temperature (see Figure 3). The full scale SNAP-8 boiler assembly procedure places the Ta tube against the outside (relative to the boiler coil diameter) radius of the 321 SS oval tube. When the assembly is heated to boiler operating temperature, the coiled diameter of the 321 SS oval tube grows more than the diameter of the tantalum tube coil due to the greater thermal expansion coefficient of the former, as mentioned previously. The differential coil diameter growth would ultimately place the tantalum tube at the midpoint of the major axis of the 321 oval tube. The differential growth in the coiled section of the boiler is accommodated by the relative movement of the two tubes, as described above, however, the differential growth in the inlet end, straight section (see Figure 3) is accommodated by a bellows. Since Test Section No. 5 was entirely straight rather than partially coiled as is the current SNAP-8 boiler conceptual design, all differential expansion between the tantalum and stainless steel structures in test section No. 5 was accommodated by a bellows at the mercury inlet end (see Figure 6). Nevertheless, the oval 321 SS tube was used so that fabrication techniques, and the effect on heat transfer of the nonuniform static NaK annular space, could be evaluated.

III. TEST PROCEDURES

A. TEST SYSTEMS

Two forced flow subscale SNAP-8 test systems were used to evaluate the various test sections. These systems were designed to provide the appropriate subscale mercury flow. They contained a primary NaK loop heat source, simulating reactor heating capacity.

Test Sections, Numbers 1 through 4, were tested in Corrosion Loop 4 (CL-4), pictorially described in Figure 7. CL-4 consists of a primary NaK loop containing a heat source coupled through the test section and a SNAP-8 simulated mercury superheater to a simulated SNAP-8 mercury-Rankine-cycle loop. The mercury loop rejects its heat through a condenser to an air-cooled circulating NaK loop. The primary NaK loop uses a resistance heater which transmits a low voltage through the NaK containment tube and the NaK to generate heat. The mercury flow passage dimensions of the four single mercury tube test sections were identical. The passage was smaller than that of a single (one of seven) full size SNAP-8 boiler tube. The operating conditions of CL-4 were set to reproduce SNAP-8 boiler operating thermodynamic conditions of mercury temperature and pressure. This required a mercury loop flow rate of 525 pounds per hour or approximately equivalent to 1/22 of the reference SNAP-8 mercury flow. The test sections evaluated represent approximately the first 4 1/2 feet of the SNAP-8 boiler. In the test section, the mercury was only partially vaporized; therefore, a workhorse 9Cr-1Mo steel boiler was included to achieve complete mercury vaporization plus an additional 150°F of superheat.

Test Section No. 5 was tested in the Seventh Scale Loop (SSL), pictorially described in Figure 8. This loop was a two-loop forced flow system. The primary NaK was heated by an electrical resistance type heater and the mercury pump provided mercury flow in excess of 5,000 pounds per hour. Approximately one-half of the mercury flow from the pump discharge was diverted through a bypass cooling loop and returned to the pump. The heater preheated the liquid mercury at the pump discharge to 500°F, the temperature at which the mercury entered the test section. In this test section, the liquid

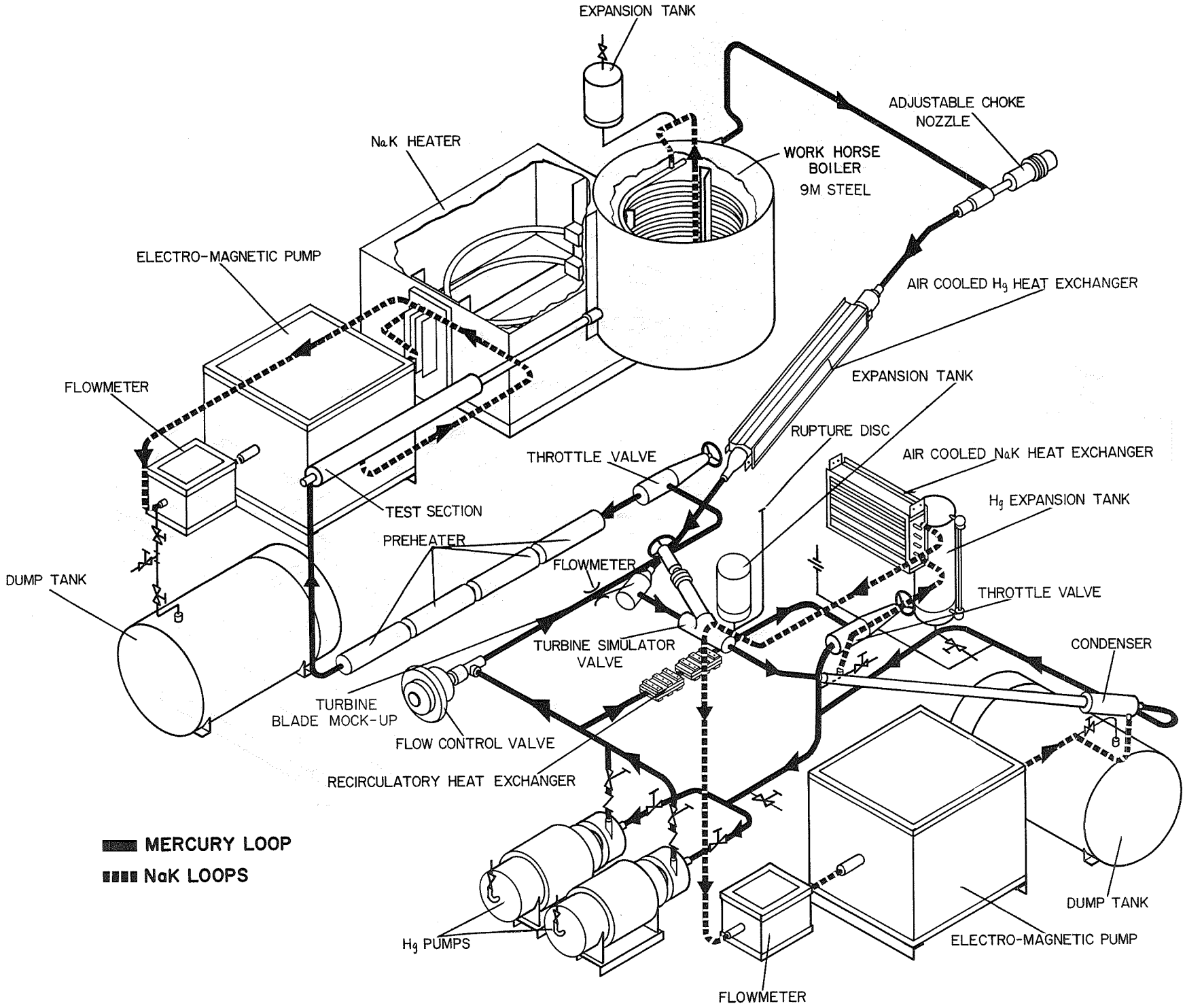


Figure 7. SNAP-8 Corrosion Loop No. 4 (CL-4) for Testing Sections No. 1, 2, 3, and 4

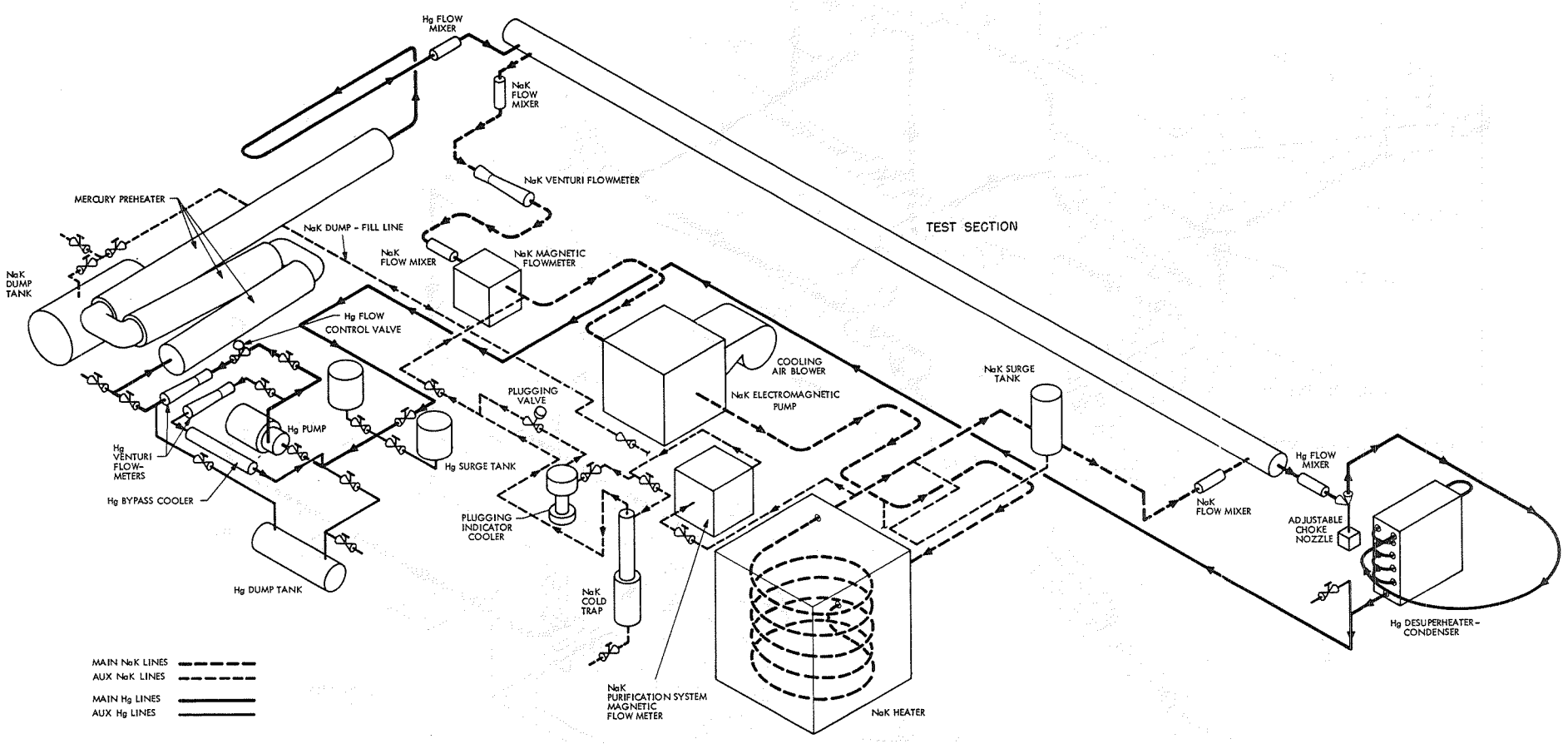


Figure 8. SNAP-8 Seventh Scale Loop (SSL) for Testing Section No. 5

mercury received sufficient thermal energy from the NaK to produce superheated mercury vapor. The vapor was then condensed in an air-cooled heat exchanger. A non-flowing NaK loop contained the inventory required to fill the static NaK annulus of test section No. 5. The static NaK system, not shown in Figure 8, contained an expansion tank mounted above the test section, a dump tank, and a static cold trap located in the cold leg between the dump tank and test section.

The mercury flow passage dimensions of the single tube test section No. 5 were identical to each of the seven mercury containment tubes in the SNAP-8 boiler. The operating conditions of the Seventh Scale Loop were set to reproduce SNAP-8 boiler operating thermodynamic conditions of mercury temperature and pressure. This required a mercury loop flow rate of 1643 pounds per hour which is approximately 1/7 of the reference SNAP-8 mercury flow.

B. OPERATING PROCEDURES

After prescribed pre-test heat transfer calibration procedures were completed on the installed test sections, the sections were operated at conditions to produce equivalent SNAP-8 mercury temperature and pressure within the test section.

During various operating periods, special tests were run to evaluate the effect of potential mercury loop contamination on the performance of the test sections. These contaminants were air and Mix-4P3E. In the SNAP-8 system, air can enter the mercury loop as a result of a failure in any component. Some new air is also introduced on any system shutdown. Mix-4P3E, the fluid used for lubrication and cooling of various SNAP-8 components, can enter the mercury loop by back diffusion through the space seals of the mercury pump or turbine, or by start-up or shut-down problems. This cross-loop diffusion can occur because the mercury and lubricant-coolant loops are exposed to a single space cavity on the space side of the seals in the SNAP-8 system.

1. Evaluation of Heat Transfer Performance

The heat transfer performance of each section was judged using several analytical constants, by comparing the predicted values with actual measurements made during test section operation. The predicted values were arrived at using heat transfer design criteria supported by previous heat transfer tests on single tubes. In this report the parameter used for indicating actual heat transfer performance is the flowing NaK side temperature drop between extremities of the test section. Again, comparison of the predicted or design value, and the actual measured value indicated whether the test section transferred heat from the flowing NaK to flowing mercury side at a rate required in the full scale SNAP-8 boiler. A value significantly below predicted, or design, would indicate that a heat transfer barrier existed.

After the performance tests were completed on each section, they were removed from the system and metallurgically evaluated to establish the cause of poor heat transfer, wherever this occurred, and whether any life-limiting reactions had occurred during the test operation. The operational test history of each of the test sections is presented in Table II.

2. Oil (Mix-4P3E) Injection Procedure

Using an ancillary injection system (see Figure 9), predetermined quantities of oil were injected at the mercury inlet end of Test Sections 2 and 3 during operation. The oil was preloaded into a line parallel to the test system liquid mercury line at the section inlet. This line was isolated from the test system by valves. Before oil was injected into the boiler, the valves were positioned so that the liquid mercury flow would be directed through the line containing the oil. The mixture was subsequently transferred into the test section.

TABLE II

TEST SECTION OPERATING HISTORY

Test Section No.	Total Operating Time Hrs.	Total Restarts	Intentional Hg-Side Contamination Test			Unanticipated Events Representing Potential Hg-Side Contamination	Chemical Cleaning
			Contaminant	Operating Time Hr.	Restarts		
1	1150	5	None	-	-	None	None
2	452	6	Mix-4P3E 2 injections each 2.5 gms	221	2	None	None
3	435	4	Mix-4P3E 2 injections each 2.5 gms	435	4	None	None
4.	2599	16	Air - 11 injections of varying quanties	985	11	Air to Hg side leak at test section outlet	Twice; first after unanticipated leak discovered. Second, after following run in which poor heat transfer performance continued.
5.	2858	11	None	-	-	1. Static NaK to Hg side leak in test section 2. Air to Hg side leak at condenser inlet	Once: After unanticipated leak No. 2.

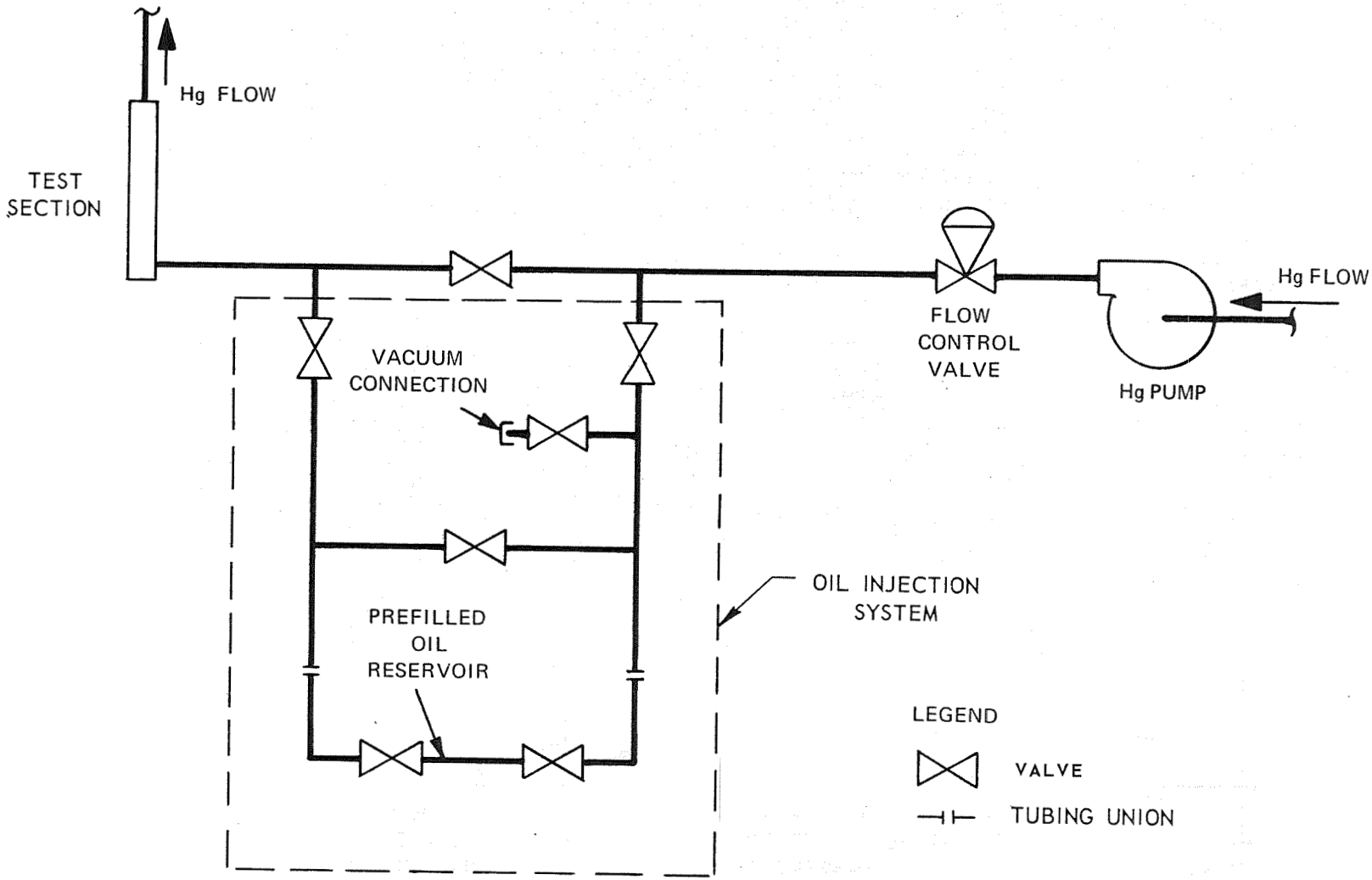


Figure 9. CL-4 Oil Injection System

3. Air Injection Procedure

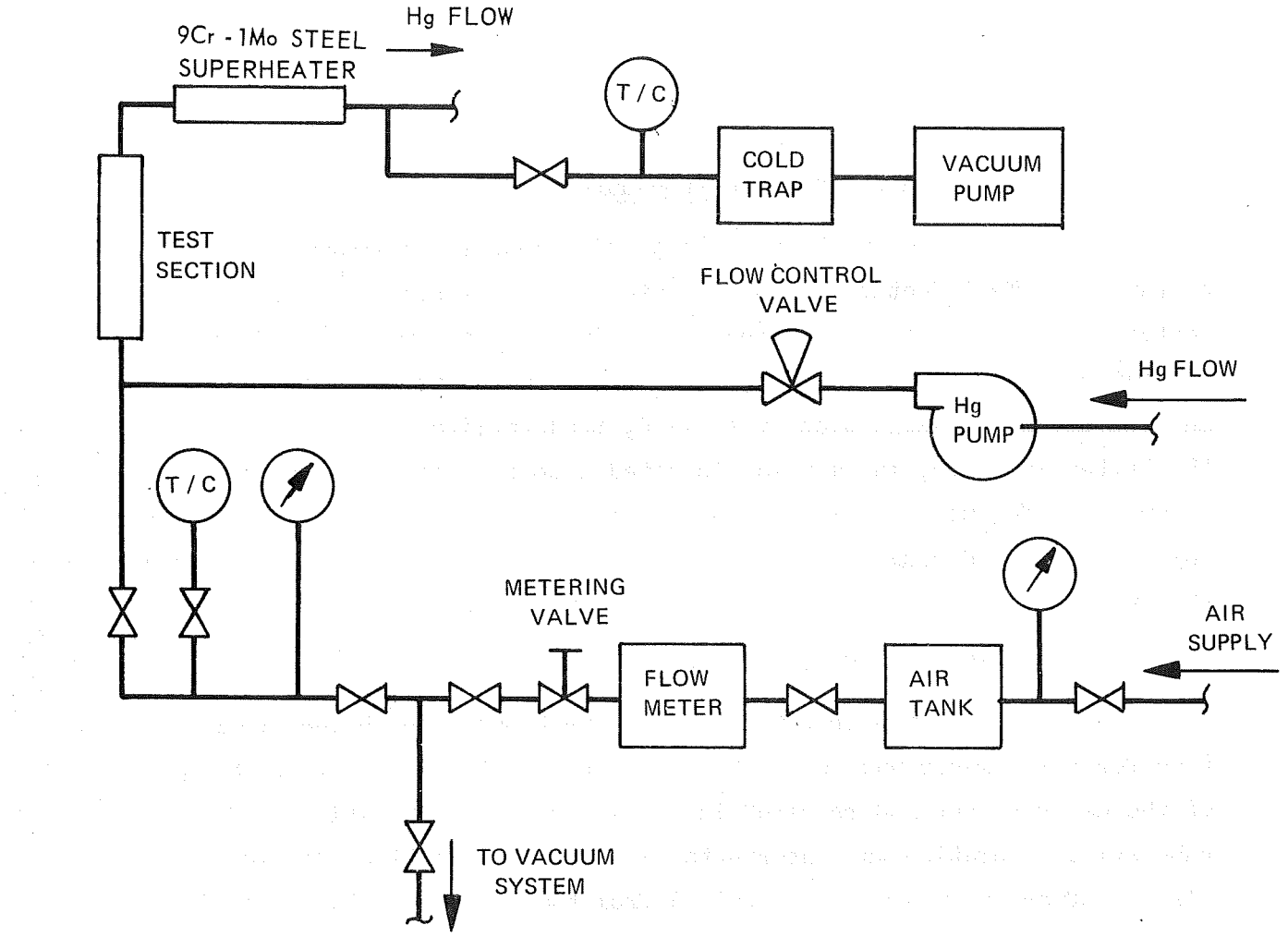
Controlled air injection into the mercury loop during operation of Test Section No. 4 simulated various possible modes of air contamination. The air was injected in either of two modes as illustrated schematically in Figure 10. The first was at a controlled rate of air flow, and the second was injection of a fixed, predetermined, volume of air. In the latter instance, the air was injected into the section either at the inlet or outlet. The test section was under an independent and continuously operating vacuum pump during air injection to insure the passage of air through the system.

C. CHEMICAL CLEANING PROCEDURE

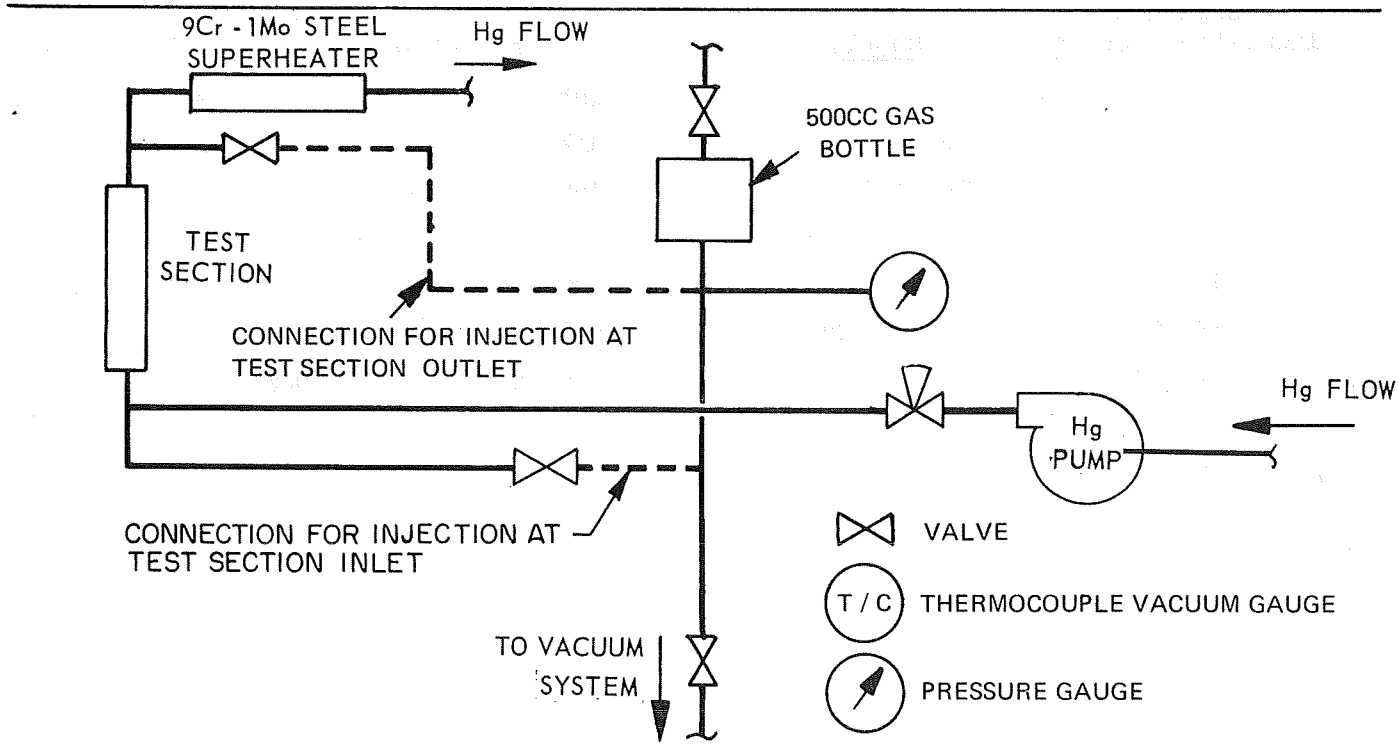
During the operation of Test Sections 4 and 5 when poor heat transfer performance was measured, it was postulated that air contamination of the mercury loop had resulted in a heat transfer inhibiting oxide on the tube wall. Operation was interrupted and the test sections were chemically cleaned after the mercury was drained from the loop using the following sequential chemical solution flushing procedure to remove this film:

Solution Identification No.	Supplier	General Description
4931	Turco Products	Inhibited Sodium Bisulfate
4338C	Turco Products	Alkaline Potassium Permanganate
4931	Turco Products	Inhibited Sodium Bisulfate
MX-12K	Cee-Bee Chemical Co.	Potassium Hydroxide
HNO_3	-	40% Nitric Acid
MX-12K	Cee-Bee Chemical Co.	Potassium Hydroxide
HNO_3/HF	-	6.2% Nitric Acid/.043% Hydrofluoric Acid

A distilled water flush was used after each solution flushing to remove residues.



(a) Controlled Rate Air Injection



(b) Fixed Volume Air Injection

Figure 10. CL-4 System for Air Injection Tests on Test Section No. 4

D. ULTRASONIC INSPECTION OF METALLURGICALLY BONDED CONCENTRIC TUBE TEST SECTIONS 3 AND 4

Ultrasonic inspection was used to quantitatively evaluate the degree of metallurgical bonding between the tantalum and 316 stainless steel tubes in the fabricated and post-test condition. Prior to inspection, the equipment was calibrated using an artificially constructed standard so that the "gate" (an adjustment limiting the search area for defects) bracketed the bond interface. In this manner, spurious indications not associated with debonding were rejected. Using a pulse echo, through-wall transmission, ultrasonic C-scan technique, the interface of the tube was inspected for debond areas which were recorded approximately 1:1, on a chart. Planimeter measurements of unbond areas on the C-scan recording provided a quantitative measure of the debonding produced by the test operation.

IV. BASIC HEAT TRANSFER PERFORMANCE

Evaluation of test data from the five test sections, which included all variations of the basic boiler tube concepts, indicated that the bonded bi-metal tube variation of concept No. 1 (see Figure 3) provided acceptable heat transfer performance. This concept was evaluated in Test Sections 3 and 4. Tube concept No. 2 evaluated in Test Section No. 5 also showed acceptable performance. The measured acceptable heat transfer performance was based on the demonstrated capability of these three test sections to operate at rated mercury outlet conditions while meeting the design criteria of flowing NaK temperature drop across the section.

Test Section No. 1 actually transferred the heat efficiently, however, post-test metallurgical evaluation indicated that the acceptable heat transfer resulted because the annular space between the unbonded tantalum and 316 stainless steel tubes was filled with flowing mercury. In effect, a double concentric mercury flow passage existed. This flow pattern defeated the prime purpose of incorporating tantalum for mercury containment, since the by-pass flow contacted the 316 stainless steel tube inner surface. Proof of by-pass flow, as an unacceptable condition, was provided by the mercury corrosion attack of the 316 stainless steel tube found in the metallographic specimens examined from Test Section No. 1 (see Figure 11).

Test Section No. 2 operated with a very low total NaK temperature drop across the test section, approximately $30^{\circ}F$, in contrast to the design value of $80^{\circ}F$. This indicated that insufficient heat was being transferred through the two (316 stainless steel and tantalum) tube walls from the flowing NaK side to the mercury. Though this section was sealed at the inlet end to prevent mercury flow through the annulus, the outlet end remained open to mercury vapor back diffusion. It was postulated that the annular space between the tubes formed at operating temperature due to the widely different thermal expansion coefficients (a calculation indicates a 0.0015 inch radial gap at $1100^{\circ}F$), backfilled with a stagnant mixture of liquid mercury and mercury vapor creating a heat transfer barrier.

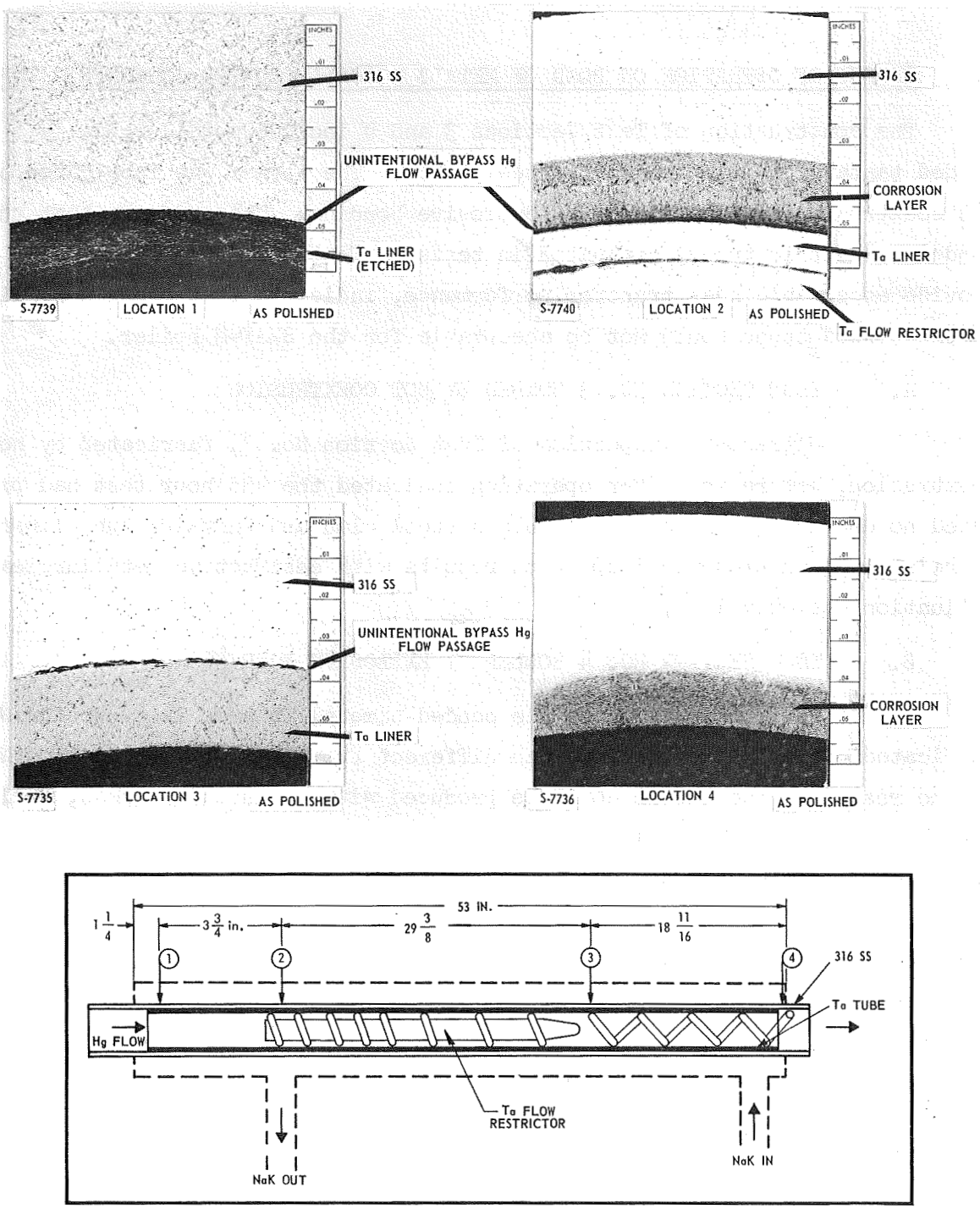


Figure 11. Hg Corrosion Attach of 316 Stainless Steel in Test Section No. 1 After 2858 Hours of Operation

V. EFFECT OF OPERATION ON BOND OF METALLURGICALLY BONDED CONCENTRIC TUBES

The construction of Test Sections 3 and 4 include metallurgically bonded tantalum to 316 stainless steel tubes. The former was fabricated by hot coextrusion and the latter by explosive bonding. The evidence that unbonded concentric tubes, evaluated in tests on Sections 1 and 2, did not provide acceptable heat transfer performance, indicated that gross debonding in the bonded tubes would not be acceptable for the SNAP-8 boiler.

A. TEST SECTION NO. 3 BONDED BY HOT COEXTRUSION

Ultrasonic inspection of Test Section No. 3, fabricated by hot coextrusion, before and after operation indicated the 435 hour test had produced no debonding of the 316 stainless steel clad and tantalum tube liner. Correlation of ultrasonic inspection results with destructive metallurgical evaluation was excellent.

B. TEST SECTION NO. 4 BONDED BY EXPLOSIVE BONDING

The construction of the bonded bimetal tube of Test Section No. 4, fabricated by explosive bonding, was different from that of Test Section No. 3 in one respect. The latter could be produced with no unbonded areas, while the former could not. The explosive bonding process required that the two tubes be separated slightly when the explosive charge was detonated to effect the bonding of the two materials. Control of this separation necessitated the strategic location of 1/16 inch diameter dimpled (inherent with explosive bonding) areas along the length of one of the tubes. Bonding did not occur at these locations of tube contact. In Test Section No. 4, the sum of the unbonded areas comprised 1.23% of the total interface area of the test section.

The section was evaluated for debonding twice during the total operating period of 2599 hours. The results are shown below:

DEBONDING OF TEST SECTION NO. 4

Operating Time Prior to Inspection (Hours)	Extent of Unbond[1] (%)
As Fabricated	1.23
369	2.22
2012	2.36

(1) Expressed as percentage of total interface area of test section.

Complete correlation of ultrasonic inspection results with destructive metallurgical evaluation could not be attained because during the air injection tests, cracking occurred in the tantalum liner from the mercury exposed surface to the tantalum-316 stainless steel interface. The inspection technique could not reliably evaluate the degree of unbond in these areas. It was also found that the tantalum cracking, and mercury corrosion of the 316 stainless steel under the tantalum cracks were not detected. A typical area containing these defects is shown in Figure 12. It was postulated that the incorporation of an additional scan using the ultrasonic shear wave technique could correct this deficiency in the inspection procedure; however, development work in this area had not been accomplished at the time this section was inspected. It would appear that regardless of the capability of the ultrasonic inspection technique for detecting these areas, this gross air contamination of the tantalum liner, in an operating SNAP-8 boiler would not be acceptable and must be avoided through reliable boiler and system design, and improved system operating procedures during ground testing.

C. LABORATORY ENVIRONMENTAL TESTS ON BONDED BIMETAL TUBES

A study was conducted on tube specimens representing each bonded tube fabrication procedure. This study was independent of the subscale loop tests on Test Sections 3 and 4. Short lengths (15 to 17 inches) of hot

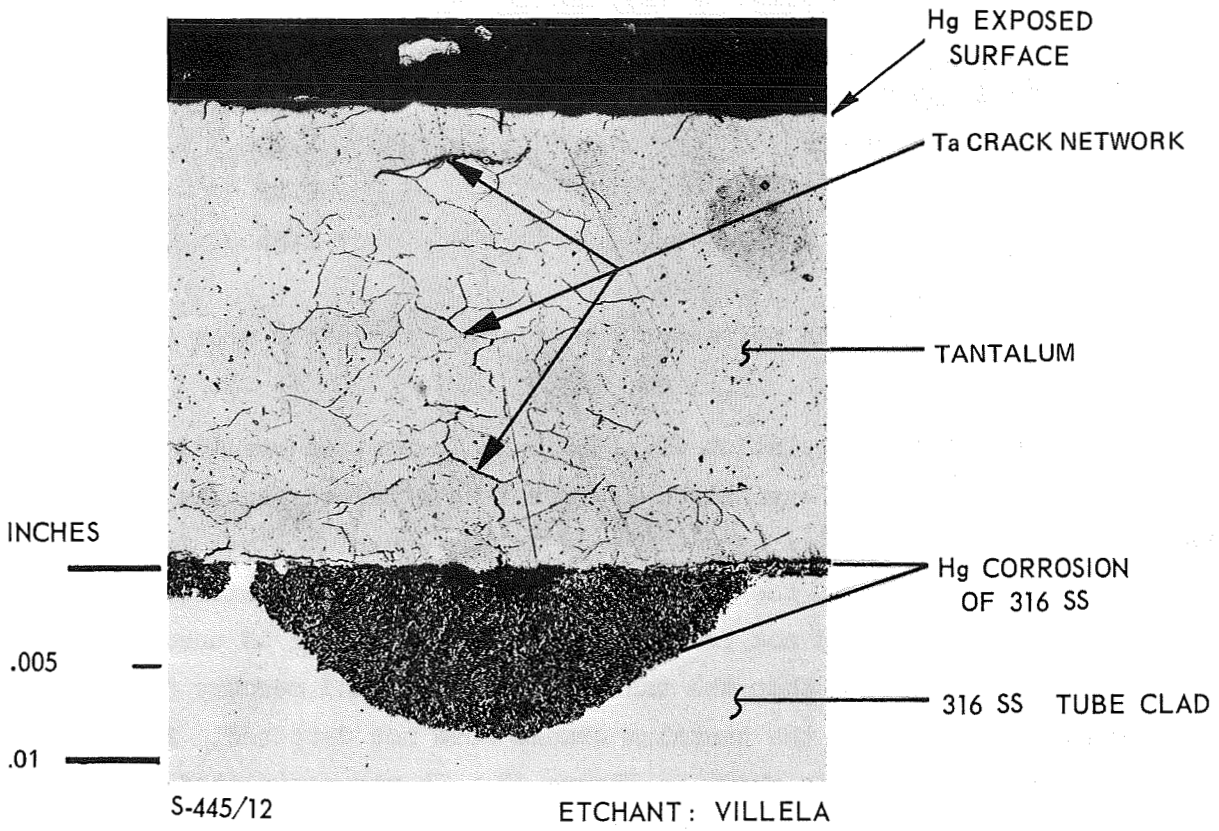


Figure 12. Tantalum Cracks and Mercury Attack of 316 Stainless Steel Bimetal Tube Clad in Test Section No. 4

coextruded and explosively bonded tubes were exposed in a vacuum furnace to a thermal environment which included cycling between 300° and 1350°F and long dwell periods at 1350°F to evaluate the debonding potential under a simulated SNAP-8 boiler operating environment. The specimens were periodically removed from the furnace and evaluated by ultrasonic inspection, utilizing both through-wall, pulse echo, and shear wave techniques. There was no debonding detected in four hot coextrusion specimens which accumulated varying exposure histories up to 3043 hours at 1350°F and 110 thermal cycles, thus confirming the non-debonding performance of Test Section No. 3.

The results of explosively bonded tube specimens (see Figure 13) indicated that the quality of the bond, and the debonding potential of pre-existing unbond areas, was related to the amount of unbond in the as-fabricated tube. For example, the specimen cut from a tube containing 40% unbond, which on a comparative basis is considered poorly bonded, exhibited a much greater tendency for growth of pre-existing unbond areas than specimens cut from tubes containing higher quantity bonds (the entire tube containing 15%, or less unbond).

It was concluded from these test data and the subscale loop tests that tantalum to 316 stainless steel bimetal tubes bonded by either hot co-extrusion or explosive bonding demonstrated the potential for providing up to 40,000 hour SNAP-8 boiler mercury containment tube life. The former method was preferred because the product contains no pre-existing sites for debond growth as does the latter. However, as long as the explosive bonding process was carefully controlled it appeared capable of producing tubes with a bond area of equal to or greater than 95%. Such a tube merits consideration for SNAP-8 boiler use in the event that the hot coextrusion process can not be developed sufficiently to produce acceptable tubes of the required 20 to 25 foot length.

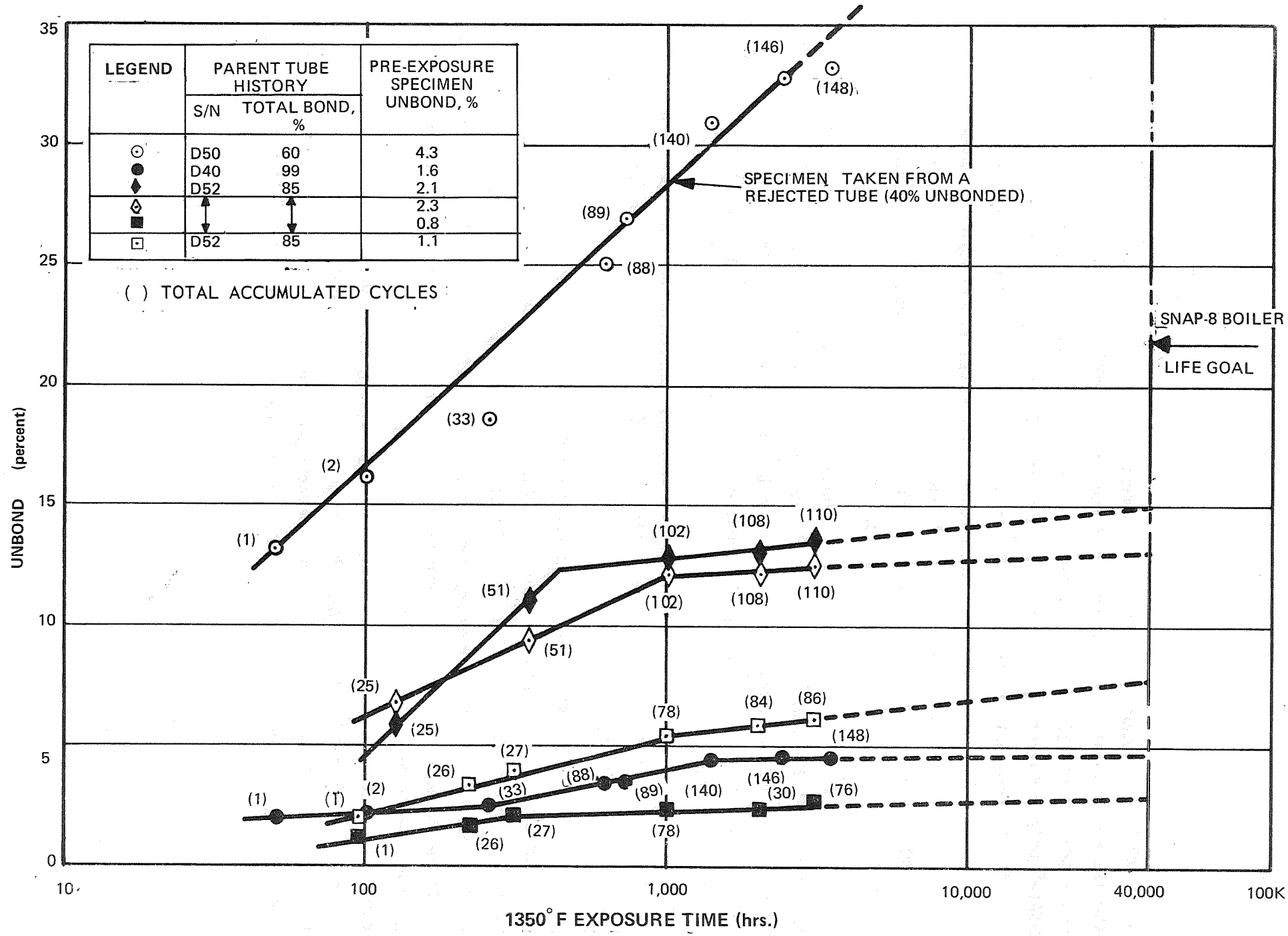


Figure 13. Debonding at 1300°F of Short Specimens Removed from 8-Foot-Long Explosively Bonded Tubes

VI. EFFECT OF MERCURY LOOP CONTAMINATION

The interstitial elements, carbon, oxygen, nitrogen, and hydrogen are readily absorbed by tantalum, the diffusion rate into the tantalum and the solubility limits being temperature dependent. The atoms of these elements diffuse between the atoms of the tantalum, residing in these interstices. At an interstitial elemental content greater than the solubility limit, as long as a continuing source is available, a reaction with the tantalum will occur forming a carbide, oxide, hydride or nitride. Until this latter reaction occurs, the presence of the interstitial elements is not visible microscopically. When the reaction does occur, the reaction product is generally observed as a precipitate in the matrix of the tantalum. To varying degrees the presence of these elements change the mechanical properties of the tantalum, generally increasing the strength and reducing the ductility.

Chemical analytical procedures were used to determine the interstitial element content of the tantalum in the test sections before and after exposure. Analysis was usually complemented by a microhardness measurement made on the tantalum because of the direct relationship between this property and interstitial element content. The procedure used in these studies was vacuum fusion analysis for gases and conductometric analysis for carbon. Pretest and post-test analytical results were compared to establish the extent of contamination resulting from operation. The effect of the contamination on heat transfer performance was established by previously described criteria of NaK temperature drop across the test section.

Air contamination at high temperature generally resulted in an increase only in oxygen content unless the amount of air was extremely large. It was found in previous studies that tantalum preferably absorbs oxygen from air until the oxygen content approaches the saturation limit. Then nitrogen would begin to diffuse into the tantalum. Oil contamination, at least the type used on the SNAP-8 program, generally, resulted in absorption of carbon, and hydrogen by the tantalum, presuming that elevated temperature oil decomposition had occurred. Associated with decomposition residue which forms as a heat transfer inhibiting film on the surface is a high carbon, and hydrogen, content.

A. ARTIFICIALLY INDUCED MERCURY LOOP CONTAMINATION

1. Mix-4P3E Injection

In the operating SNAP-8 system, some oil will diffuse from the lubricant-coolant loop of the system into the mercury loop through the turbine and mercury pump space seals. The two loops are exposed to a common chamber on the space side of the seals. Two potentially detrimental effects of oil in the mercury loop are postulated. Both result from thermal decomposition of the oil when exposed to the high SNAP-8 boiler temperature of greater than 1100°F. The critical temperature range for oil decomposition is 800° to 850°F. First, the decomposition forms elemental carbon and hydrogen. An excess of these elements in the tantalum can cause severe hardening and embrittlement of this material. Second, oil decomposition results in formation of a carbon residue and/or varnish which, if deposited as a coating on the tube wall, acts as a heat transfer inhibiting layer.

Test Sections, No. 2 and 3, were used to evaluate the effect of the presence of Mix-4P3E oil in the test section during operation. Mix-4P3E oil was injected into the mercury system at the test section inlet and the mercury flow transported the oil into the section. The quantity injected at one time was 2.5 grams. This amount represents the equivalent (based on scaling factor) total quantity of oil entering one tube of the seven tube SNAP-8 system boiler during 10,000 hours of continuous operation.

Test Section No. 2 did not represent a valid heat transfer test because the effect of the oil injection on performance could not be established with certainty due to the inherent poor heat transfer of the test section design.

During steady state operation of Test Section No. 3, prior to oil injection, the test section exhibited design-predicted heat transfer performance as indicated by a flowing NaK temperature drop across the section of 110°F. Two 2.5 gram charges of Mix-4P3E oil were individually injected into test section No. 3. The first injection was made after 235 hours of operation and no change in the NaK temperature drop occurred with

this first injection. The mercury flow was then stopped, simulating mercury system shut-down; however, the NaK flow was continued at 1100°F. Mercury flow was restarted after 16 hours and no change in heat transfer performance was indicated as seen by a continuous NaK ΔT of 110°F. A second 2.5 grams of Mix-4P3E was injected into the mercury loop and the above procedure repeated. During the operating period immediately following injection, no change occurred. After shutdown and restart, the heat transfer performance was degraded from the condition which existed prior to the shutdown. The NaK temperature drop across the test section had decreased from 110°to 37°F. Operation of the test section was continued without making any changes in the loop operating parameters. During the following 36 hours, the performance of the test section returned to its original heat transfer level. A second shutdown and restart was performed with no resultant heat transfer performance degradation.

Post-test visual examination of the mercury tube, after it was cut longitudinally, revealed a black deposit in the mercury flow channel, at the mercury inlet end of the tube. However, post-test chemical analysis indicated that no detrimental effect on the tantalum resulted from the Mix-4P3E decomposition. This test section also showed no significant increase of carbon, or hydrogen in the tantalum liner.

2. Postulated Theory of Performance Degradation Due to Mix-4P3E

The following mechanism was postulated to explain the performance of the test section resulting from the Mix-4P3E oil injection. The viscosity of the oil was markedly decreased by the increased exposure temperature. Furthermore, at 400°F and above, the oil completely wets the mercury and a mechanical mixture of oil and mercury exists in the flowing mercury. For the reaction below 400°F, the oil exists as spherical droplets in the mercury. To further describe the phenomenon above 400°F, if a small quantity of oil is dropped on a free surface of mercury, a very thin film forms over the entire mercury surface; whereas at 75°F the oil remains on the surface as a drop. The contact angle between the drop and the mercury surface would be less than 90

degrees in the latter case. The test section inlet temperature was approximately 500°F, therefore, the former condition would exist. This situation, in combination with the ability of tantalum to become wet by mercury (i.e., formation of a surface adherent thin liquid mercury film) in the all liquid mercury portion of the test section, and the turbulent flow of mercury through the test section acted in concert to prevent contact of the oil with the tantalum tube wall. If contact had occurred, the oil would have been decomposed since the tantalum tube wall temperature (varying up to 1300°F) exceeded the bulk decomposition temperature of the oil, 800° to 850°F.

When the mercury flow stopped, the mercury cooled and the mercury vapor in the system condensed, a vacuum in the non-filled portion of the loop was created. This space was subsequently filled with mercury and non-decomposed oil vapors and oil decomposition gases, primarily hydrogen, CO_2, CO and methane, to satisfy the equilibrium partial pressures of these constituents throughout the system. When the non-decomposed oil vapors contacted the tantalum tube wall, which was maintained at 1100°F by the continuously flowing NaK, decomposition resulted producing a carbon film on the tantalum surface. Similarly a film was produced as a result of a reaction between the tantalum tube wall and the oil decomposition gases. In effect, the test section tube wall gettered most or all of the oil vapor and decomposition gases in the system.

Additional analytical and experimental work is being performed at Geoscience Limited, Solana Beach, California under the auspices of NASA's Lewis Research Center in an effort to gain a complete understanding of the phenomena.

In the Mix-4P3E oil injection test on Test Section No. 3, the first 2.5 grams of oil injected did not affect the performance of the section for several postulated reasons.

. As long as the test system operation continued after the injection, the oil could not contact the tube wall and no heat transfer inhibiting decomposition film could be deposited at the mercury inlet end of the boiler which is the most critical heat transfer area.

. Upon shutdown, the film was deposited but the total amount of oil available and/or the total down time was not adequate for a film buildup to reach that critical thickness which would affect heat transfer performance during the subsequent operating period.

The 2nd oil injection of 2.5 grams did not cause heat transfer performance degradation as long as system operation continued, because the conditions of operation prevented the oil from contacting the tube surface. However, when the mercury flow was stopped, two sources of surface film buildup were now possible.

. The undecomposed oil remaining in the system from the first injection of 2.5 grams.

. The second injection of 2.5 grams.

Based on degraded heat transfer performance after mercury system restart following the second injection, it appeared that the film deposited during shutdown following the first injection of 2.5 grams plus that contributed by one or both of the above sources produced a critical amount of decomposed oil film thickness on the tantalum.

During the subsequent continuous operating period of 36 hours the flowing mercury produced a scrubbing, self-cleaning action and a sufficient amount, though not all as evidenced by the remaining black layer seen during post-test inspection, of this film was mechanically removed from the tube wall. The original heat transfer performance, indicated by a flowing NaK temperature drop of 110^{o}F through the test section, was restored.

After this recovery was completed, the mercury loop shut down and the restart procedure was repeated with no detrimental consequence on heat transfer performance indicating that a sufficient portion of the total 5 grams of oil injected into the system had been decomposed during the previous two shutdowns. Therefore, decomposition of any remaining oil did not produce a critical film thickness on the tube wall to affect heat transfer performance.

3. Air Injection

Test Section No. 4 operated with an air injection system which simulated air leakage into the mercury system during a PCS system failure. Data analysis indicated that a reaction between the tantalum tube and some presently unknown amount of air in the system would degrade heat transfer performance and change the mechanical properties of the tantalum. A detrimental reduction of tantalum ductility could result. The mechanical properties were changed as a result of interstitial absorption by the tantalum of the oxygen and nitrogen from the gas in the system.

Using the technique described in Section III-B-3, eleven air injections were made (see Table III). The injections were made into a non-flowing mercury system but with flowing NaK in the primary loop, which produced a high temperature tantalum tube wall varying between 1000° and 1200°F. The high temperature of the tantalum simulated the SNAP-8 operating temperature which facilitated diffusion of the gases into the tantalum tube wall. The performance of Test Section 4 during these experiments verified the detrimental effects of air on heat transfer performance of tantalum tubes. Based on these tests and visual observance of disassembled test sections after confirmed air leakage due to mercury loop failures (described below), it was apparent that the cause of the reduced heat transfer performance was a tantalum oxide surface film produced by the reaction of tantalum with air at elevated temperature. This film forms very rapidly above 400°F.

Quantitative analysis of the test data from air injection into Test Section 4 was not possible because it was established, as part of post-test evaluation, that a NaK loop to mercury loop leak probably existed during most, if not all, of the test series. The leak was confirmed by detection of NaK in the mercury inventory. It had been established by previous experiments that alkali metal additions to mercury would significantly enhance heat transfer performance by facilitating removal of metal surface oxides and consequently wetting of the wall by the mercury. Therefore, the air injection test results were, if anything, conservative in their indication of the detrimental effect of air on the heat transfer performance of tantalum mercury containment tubes in the SNAP-8 boiler. Had there been no NaK in the mercury, the heat transfer performance of the test section would probably have been much more adversely affected by the air injection.

TABLE III

AIR INJECTION INTO TEST SECTION NO. 4

Air Injection Test No.	Accumulated Hrs of Air Exposure Prior to Injection	Ta Tube Temp °F	Air Injection		After Air Injection[1] NaK ΔT °F	Injection[2] Procedure
			Rate cc/min	Total Quant. cc/(STP)		
1	0	1000	0.5	1.4	120	A
2	25	1200	0.5	1.4	100	B
3	210	1200	0.5	1.4	60[3]	B
4	235	1200	0.5	1.4	120	B
5[4]	255	1100	0.5	1.4	100	B
6[4]	280	1100	2.5	69.6	120	B
7	300	1200	3.5	195	100	B
8	440	1100	(5)	15.8	80	C
9	530	1140	(6)	31.6	35	B
10	625	1200	(7)	2785	71	B
11	815	1200	1.5	250	50	D

Notes:

(1) Base Point: NaK $\Delta T = 120^{\circ}F$.

(2) The following four different modes of air injection were used in an attempt to simulate four potential mercury loop air leak conditions.
- (A) Air injection at slow controlled rate into test section inlet.
- (B) A fixed volume of air, 500 ml, was injected at the test section inlet.
- (C) Same as "A" above except that injection point was at the TSE mercury superheat outlet.
- (D) Same as "B" above except that injection point was at the test section outlet.

(3) The third injection was made at a very short operating interval after the second injection. The NaK ΔT base point was $100^{\circ}F$.

(4) The mercury loop was not evacuated prior to restart for these tests only.

(5) A single quantity of 500 ml of gas was injected. The 500 ml was a mixture of air and argon. The relative proportions were established by partial pressures, i.e., 1 inch mercury partial pressure of air in 74 psig argon.

(6) A single quantity of 500 ml of gas was injected. The 500 ml was a mixture of air and argon. The relative proportions were installed by partial pressures, i.e., 2 inches mercury partial pressure of air in 74 psig argon.

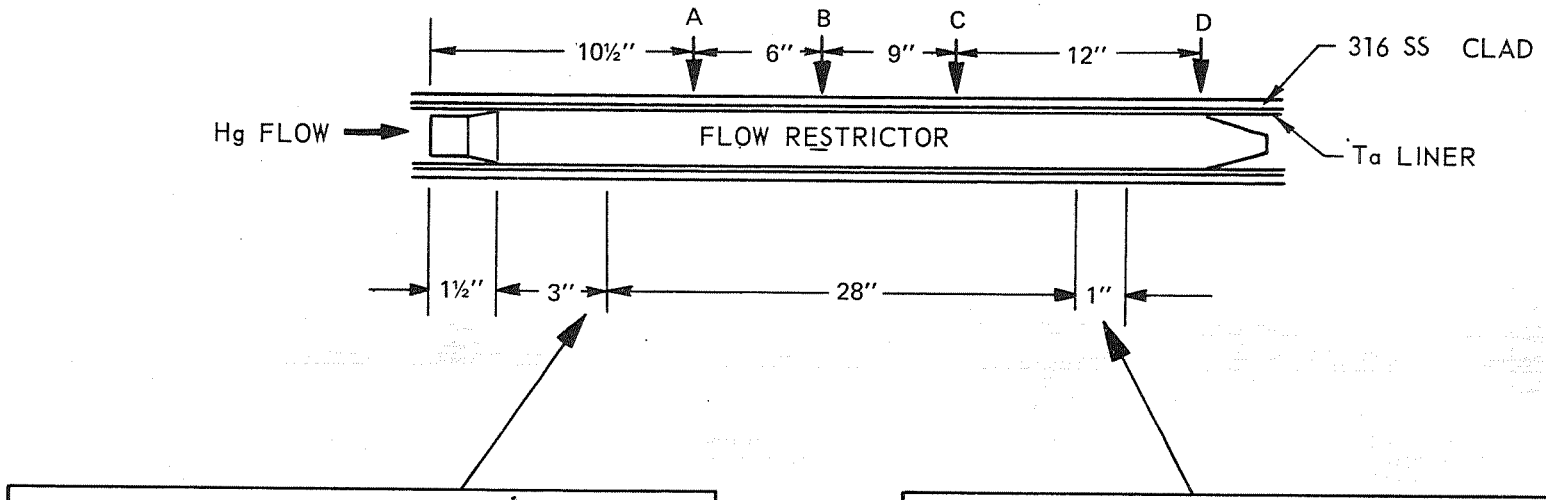
(7) 500 ml bottle filled with pure air at 75 psig was injected.

A minor air-tantalum interaction was evident in tantalum samples from the plug insert region, and was characterized by: an increase in the oxygen from a pretest value of 85 ppm to 740 ppm in the tantalum liner, the development of a subsurface oxide, approximately one mil deep in the tantalum liner and tantalum plug, and by an increase in the tantalum ID microhardness and plug OD microhardness (see Figure 14). A major tantalum-air interaction was evident in the area immediately downstream of the plug insert region and was characterized by an extensive cracking in the tantalum liner (see Figure 15), and reduction of the wall thickness of the tantalum liner.

Cracks were evident in the liner ID, OD, and center, and several extended from the tantalum liner ID to the OD. Microhardness measurements taken adjacent to the cracks were approximately equivalent to microhardness measurements taken in the uncracked area. When tantalum-air interaction occurred, the microhardness increased and when the diffused oxygen concentration exceeded the saturation solubility limit for the exposure temperature, tantalum oxide precipitation occurred in the grain boundaries and on certain crystallographic planes. One explanation for the cracking would be that it was caused by the combined effect of thermal cycling stresses and reduced ductility of the tantalum due to the air-tantalum interaction.

The tantalum liner wall thickness in the plug exit region varied from 14 to 19.5 mils. The variation of the "as-received" sample was 18.8 to 19.5 mils. No particular correlation was found between the cracking and the wall thinning. For example, ID to OD cracking was found in a thin wall and normal wall thickness areas. The tantalum wall thinning was probably the result of ID oxidation of the tantalum liner followed by spalling of the oxide layer.

Stainless steel leaching was evident in the plug exit region. The mercury attacked the stainless steel internal diameter surface and leached out certain areas (see Figure 15). The attack occurred in areas adjacent to



Parts Per Million of	C	O	H	N
Flow Restrictor	40	22	4	30
Ta Liner	40	740	6	30

Parts Per Million of	C	O	H	N
Flow Restrictor	25	50	3	20
Ta Liner	35	390	4	30

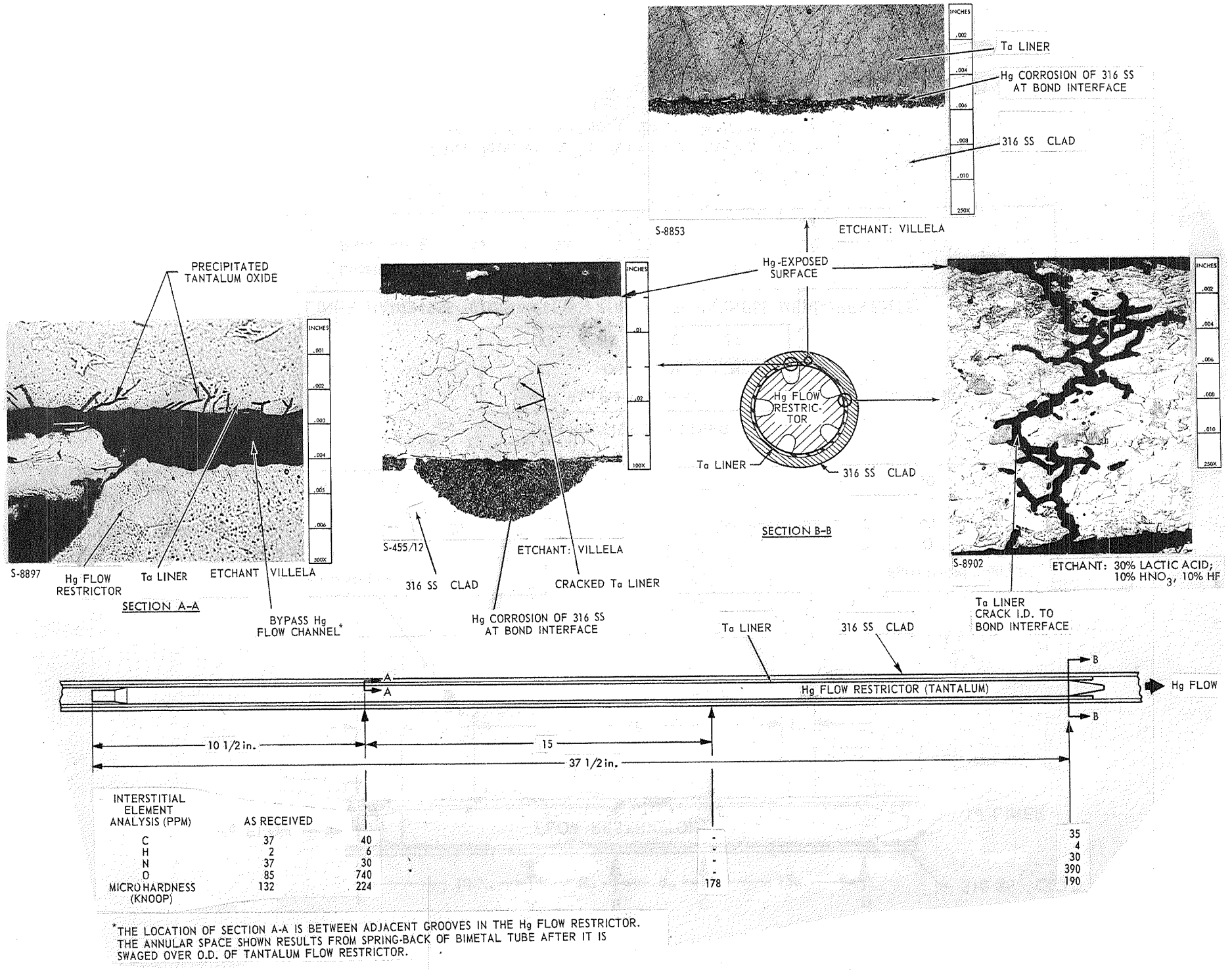
AS RECEIVED Ta LINER

Part Per Million of			
C	O	H	N
37	85	2	37

LINER HARDNESS (CONVERTED FROM MICROHARNESS MEASUREMENTS)

Location	A	B	C	D	As Received
Rockwell B	99	94	101	95	75

Figure 14. Interstitial Analysis of Tantalum Liner and Mercury Flow Resistor and Liner Hardness Test Section No. 4



INTERSTITIAL ELEMENT ANALYSIS (PPM)	AS RECEIVED				
C	37	40	-	35
H	2	6	-	4
N	37	30	-	30
O	85	740	-	390
MICRO HARDNESS (KNOOP)	132	224	178	190

*THE LOCATION OF SECTION A-A IS BETWEEN ADJACENT GROOVES IN THE Hg FLOW RESTRICTOR. THE ANNULAR SPACE SHOWN RESULTS FROM SPRING-BACK OF BIMETAL TUBE AFTER IT IS SWAGED OVER O.D. OF TANTALUM FLOW RESTRICTOR.

Figure 15. Effect of Air Injection on Tantalum 316 SS Bonded Bimetal Tube in Test Section No. 4

cracks that formed at the tantalum to stainless steel interface. Apparently the mercury diffused through the network of cracks in the tantalum liner ultimately contacting the 316 stainless steel clad at the tantalum/316 stainless steel interface. Subsequent leaching-type corrosion, typical of mercury attack of 316 stainless steel, occurred. Based on this corrosion mechanism, and the anticipated rate of attack established by other studies, the Type 316 stainless steel tube was estimated to be under attack for 610 hours.

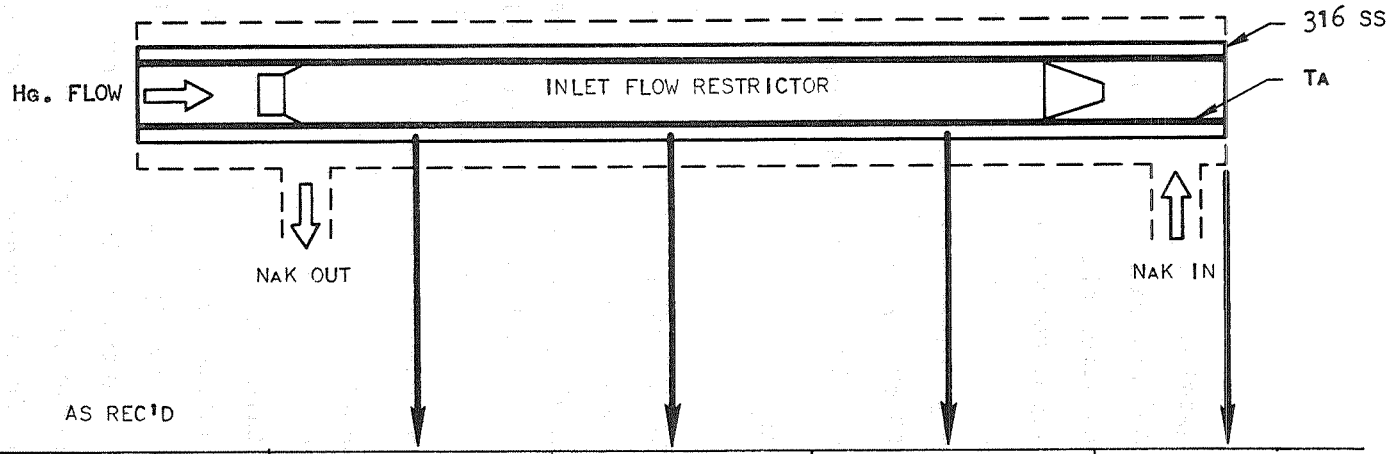
B. Hg LOOP CONTAMINATION CAUSED AFTER TEST SYSTEM FAILURE

1. Test Sections 1, 2 and 3

Air was not intentially injected into these sections and there was no obvious system failure which could have resulted in air contamination. However, interstitial analysis of the tantalum indicated that an undetected source of air had contaminated the tantalum tubes in each of these test sections. An increase in hardness as a result of section operation, determined by microhardness measurements, confirmed the chemical analysis results. The pre- and post-test comparative data were as shown in Figure 16.

The interstitial analysis and microhardness checks indicated that the tantalum in each section picked up oxygen and nitrogen, especially near the mercury inlet end of the tantalum liner. Some increase in strength and a reduction of ductility of the tantalum could be expected from the increase in oxygen and nitrogen content but it is likely this would not significantly affect the corrosion resistance of the tantalum liner. The most likely sources of the nitrogen and oxygen were air contamination during the mercury loop out-gassing procedure performed at startup or in-leakage of air when, during mercury loop shut-down, a subatmospheric pressure resulted from mercury vapor condensation.

The data indicated that there was no internal contamination of tantalum from the thermal decomposition of Mix-4P3E in Test Sections No. 2 and 3, which were subjected to oil injection tests. Neither was there oil contamination from the oil diffusion pumps used prior to loop startup. The tantalum liner increased primarily in oxygen and nitrogen content with no carbon



		AS REC'D														
	TEST SECTION NO.	1	2	3	1	2	3	1	2	3	1	2	3	1	2	3
INTERSTITIAL ANALYSIS (PPM)	C	50	50	45	40	65	45	-	55	40	20	30	60	95	30	55
	H	7	7	17	5.5	8	2.5	-	2.5	1.5	5.5	3	2	5.5	3	2
	N	25	25	40	245	75	50	-	285	15	115	40	35	70	15	35
	O	20	20	40	145	115	80	-	340	20	50	160	35	45	75	55
HARDNESS, ROCKWELL B (CONVERTED FROM MICRO-HARDNESS MEASUREMENTS		97	97	91	93	105	92	88	102	94	-	100	92	88	100	93

Figure 16. Interstitial Analysis and Hardness of Tantalum Tube in Sections 1, 2 and 3

or hydrogen pickup noted. Any increase in the latter two elements would have been measured if oil had inadvertently entered the loop. It appeared that the breakdown of Mix-4P3E injected into Test Sections No. 2 and 3 did not materially contribute to tantalum contamination and that in-leakage of air during startup or test operation was the main source of interstitial buildup. There was an indication of much less air contamination during the test of Section No. 3 when compared with the first two test sections.

2. Test Section No. 4

After 165 hours of Test Section No. 4 operation, a momentary commercial power loss shut the loop down. The reference operating conditions were restored in approximately 5 minutes, and it was noted at this point that the heat transfer performance of the section had degraded. This was indicated by a sudden reduction in the NaK temperature drop. After 99 hours of additional operation with the continued poor performance, a mercury leak to air was found in the test section outlet pressure transducer and the loop was shut down for repair.

Upon completion of repairs, the test section was restarted and the heat transfer performance of the boiler was still subnormal. Operation continued for an additional 105 hours with no change in performance. At this time the test was discontinued and the test section was removed from the system. An interstitial element analysis was performed on a segment of the tantalum tube. The stainless steel clad was dissolved with aqua regia and the tantalum analyzed for interstitial elements of C, H, N, and O. The results were as follows:

	Content of Tantalum (PPM)	
Element	Post-test	Pretest
C	65	37
H	16.5	2
N	215	37
O	365	85

The sources of the above elements were air (O and N) and oil (C and H). Air could have been introduced through a system leak, and oil could enter by back diffusion from the oil diffusion pump used in system evacuation during the test of this and previous sections.

The Test Section was chemically cleaned to remove any heat transfer inhibiting surface films, using the solutions described in Section III-C. The residues present in the test section were collected on a filter incorporated in the cleaning (forced-fluid flush) system. To prevent mixing of residues from the cleaning system, with those from the test section, a second filter was placed at the end of the test section through which the fluid entered. The test section residue contained 10.2% C, and 1.49% H, which was indicative of oil decomposition on the tantalum tube surface.

A transverse sample of the bimetal tube was removed from the inlet end and evaluated metallographically. No subsurface precipitated tantalum oxides were visible in the microstructure. However, a microhardness traverse through the tantalum liner indicated converted hardnesses of Rockwell B 88, 96, and 107 at the stainless steel interface, at the center of the tantalum wall, and at the mercury exposed inner surface, respectively. In comparing a pretest hardness of 70, and considering the marked hardness gradation, there was an obvious indication of hardening as a result of exposure to some contaminating environment on the mercury side of the bimetal tube.

After cleaning Test Section No. 4 was re-installed in the loop for continued testing. Upon restart, heat transfer performance of the test section was as poor as it was immediately prior to the previous shutdown and cleaning, and remained so through 218 hours of additional operation. The NaK temperature drop was approximately 50°F compared to 130°F during the first run. The test was terminated and the section was removed for recleaning. When the section was removed from the test loop an abundance of white powder was discovered inside the tube at the mercury outlet end. An X-ray diffraction analysis identified this substance to be primarily Ta_2O_5. An insignificant amount of C, H and N were also present in the tube. It was apparent that

during the run following the first cleaning operation an undetected air leak existed in the system downstream of the test section mercury outlet. After recleaning, using the same procedure as was used the previous time, and resumption of test operations rated heat transfer performance was produced.

3. Test Section No. 5

a. Operational History

Test Section No. 5 operated for a total of 2858 hours, which included 12 restarts. During a majority of the restarts, the initial heat transfer performance was poor. However, performance improved with continued operation and generally rated performance was achieved within 50 to 175 hours after each startup.

Approximately half way through the testing of this section, the SNAP-8 project adopted a more stringent procedure for starting any system containing a tantalum mercury containment component to avoid undetected mercury loop leaks which would produce air contamination of the tantalum tube. Prior to the procedure change, during system start the mercury loop was evacuated to approximately 10 microns maximum, and the primary NaK loop inventory was heated to 1300°F. The NaK was flowing through the NaK side of the test section, and therefore heated the tantalum which aided in mercury loop outgassing. If an air leak existed, the heated tantalum absorbed the gas and became contaminated.

The new system start procedure required that during heat-up of the NaK primary loop, prior to heating to above 400°F (critical temperature for surface tantalum oxide formation), the integrity of the loop would be checked by performing a pressure-rise rate test. This was accomplished by isolating the vacuum pump from the loop by valving and measuring the rate of pressure buildup in the loop. This test indicated whether the loop was leak-tight. The acceptance criteria was that within 24 hours the rate of pressure increase must be reduced to a point which showed an asymptotic relationship to some low value, obviously not indicative of an air leak but rather due to a very slow outgassing of the loop materials toward an equalized internal pressure

condition. The use of this procedure during one restart checkout resulted in the discovery, and replacement, of a leaky valve in the vacuum take-off line downstream of the condenser. This valve could have been leaking air into the system during the entire previous test history causing tantalum oxidation and subsequent conditioning problems experienced during every shutdown.

A second operating procedure change was made to protect against undetected, but small, Hg loop leaks of this type. Originally during the system shutdown, there existed a period of time, after mercury flow ceased, when the mercury loop pressure dropped below atmospheric due to mercury vapor condensation, thus accentuating the potential for air introduction. Since the primary NaK loop continued circulating, the tantalum remained above the critical surface oxidation temperature of 400°F. The procedure change required an argon back-fill of the Hg loop upon the cessation of Hg flow to insure an internal pressure above atmospheric at all times. Thus, system protection against air introduction through an undetected leak was maximized.

Two running periods followed the above repair and changed operational procedures. The test section performed poorly immediately upon each restart, however, an improvement was noted with operating time. Vacuum decay tests and a helium leak check following this second period disclosed a leaking pressure transducer fitting on the condenser inlet line. This fitting was tightened and pressure decay tests verified the absence of leaks. The test section was restarted and again the poor performance was observed. Apparently the leak had been so large that the changed system operating procedures had not been sufficient to protect against boiler contamination. The heat transfer performance improved steadily over the next 70 hours of operation, although rated performance was never achieved. At this time, four rapid power losses occurred and three hours later the heat transfer performance had degraded rapidly. During the next four days the section was operated at reduced mercury flow in an unsuccessful effort to improve boiler performance. After a total operating time of 164 hours, large amounts of mercury were discovered beneath the mercury condenser and the test section. The test operation was manually shut down and a leak was located at the junction where an 1/8 inch pressure transducer sensing line was welded to the condenser inlet line.

During this shut-down period the test section was cleaned using the chemical solutions described in Section III-C. to remove any heat transfer inhibiting oxide films which may have formed during the previous operating periods with air leaks. After system repair and test section cleaning, the test was resumed. Immediate rated heat transfer performance of the section was achieved at startup and continued through the balance of the run which lasted for 165 hours.

The operational history, and heat transfer performance of this test section indicated the extreme criticality of having good control over the integrity of the system. Every effort must be made in operating a high temperature system containing heat transfer surfaces of tantalum to keep the system free of leaks. Any air leak results in a reaction with the tantalum producing a heat transfer inhibiting tantalum oxide surface film.

b. Post Test Evaluation

Post-test evaluation by microhardness measurement and interstitial element analysis, see Table IV, after the final 165 hour operating period indicated tantalum tube contamination apparently had occurred, notably by carbon and oxygen. However it obviously was not sufficient to detrimentally affect the satisfactory heat transfer performance of this last test run.

TABLE IV

INTERSTITIAL ELEMENT CONTENT OF Hg EXPOSED SIDE OF THE TANTALUM TUBE IN TEST SECTION NO. 5

	Interstitial Analysis (PPM)					
		Distance from Hg Inlet (inch)				
Element	Pretest	17	58	114	177	282
Oxygen	20	40	45	38	65	65
Nitrogen	39	46	35	24	47	10
Hydrogen	11.5	17	8	8	7	6
Carbon	10.5	75	55	62	90	200

An interstitial analysis of the tantalum mercury inlet flow restrictor and tantalum-10% W turbulator wire of the test section also showed apparent marked carbon contamination, notably at the mercury inlet end of the flow restrictor and the mercury outlet end of the wire (see Table V).

TABLE V

TANTALUM PLUG AND TANTALUM-10% TUNGSTEN WIRE INTERSTITIAL CHEMICAL ANALYSIS OF TEST SECTION NO. 5

Sample Location Inches from Hg Inlet	Element (ppm)			
	O_2	N_2	H_2	C
Flow Restrictor				
13-1/2	84	28	9	1010
45-1/4	81	19	8	70
60	71	35	8	83
Wire				
64	100	11	8	75
355	335		4	340

A surface coating was noticed on the mercury exposed surface of all tantalum components in the test section. These included the inside wall of the tantalum tube, the outside surface of the tantalum mercury flow restrictor, at the mercury inlet, and at the outside surface of the tantalum alloy (Ta-10%W) turbulator wire downstream of the flow restrictor. The coating appeared as a mixture of microstructural constituents when viewed at high magnification. Of particular significance was the presence of a similar coating on the as-received tantalum tube (see Figure 17). An investigation

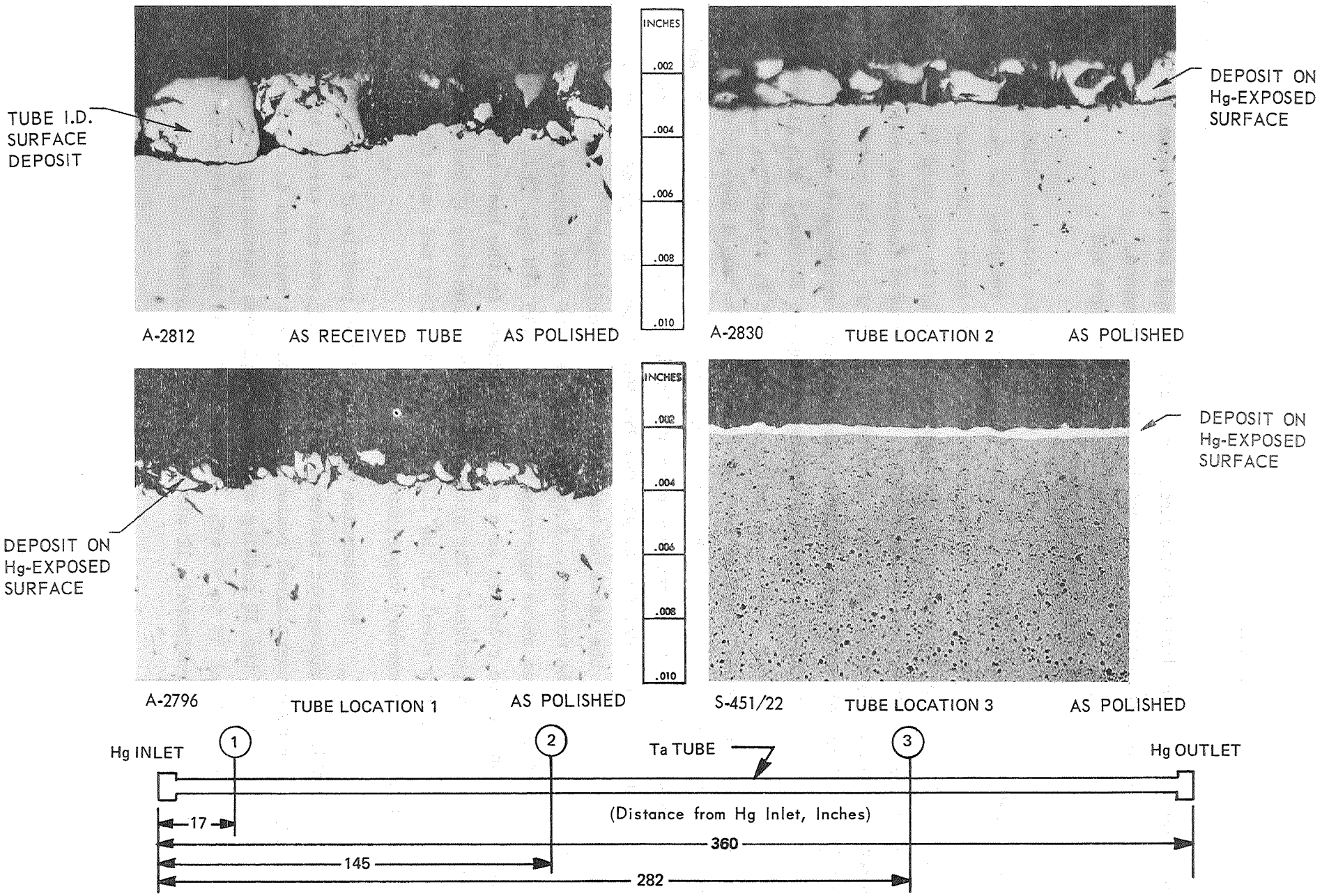


Figure 17. Coating on Hg Exposed Surfaces of Ta Tube in Test Section No. 5

was conducted to evaluate the nature of the coating and perhaps establish a reason for no adverse effect on heat transfer performance.

(1) Surface Film Physical Structure and Chemical Composition

The test section tantalum tube internal diameter (ID) coating thickness varied along the tube length, reaching the maximum of .003 inches at a point 60 inches from the mercury inlet. Emission spectrograph, X-ray fluorescence, and microprobe analysis was used to establish the composition of the coating. The unexposed (as-received) tube contained primarily Al, with small amounts of Mg and Fe in the surface residue. These elements originated as some unknown contaminant during the tube fabrication process or perhaps during subsequent storage. The Al and Mg elements are highly soluble in mercury, even at 75°F, therefore, it is postulated that removal through test section operation would have occurred. The post-test surface deposit chemistry indicated approximately 43 wt% Ni and approximately 37 wt% Ag as primary elements in the film. Less than 1% each of the following, Cl, Fe, Cr, K, Al, Ca, Pu, Mn, Zr and Nb were also found. The silver found on the tantalum tube ID probably originated as an impurity in the mercury. A typical analysis of pure mercury prior to loading in the system shows approximately 0.1 to 0.15 PPM Ag. This element will drop out of the solution as a solid precipitate in the heat transfer region as the Hg vaporizes. The nickel on the tantalum tube ID was leached out of the stainless steel in the liquid leg of the loop and mass transferred to the tantalum as mercury vaporization occurred.

To determine the composition profile of the tantalum tube ID coating, a microprobe traverse for Ni, and Ag was run across the coating. For some unexplained reason, silver was not detected in the traverse. Nickel was found in the ID coating with a semi-plateau indicating a probable metallic compound, at 35 to 50 wt%. Nickel diffusion into the tantalum was found extending approximately 12 microns below the surface.

(2) Surface Coating Structure

The surface structure analyses were performed with the aid of X-ray diffraction, and electron diffraction analysis. Three compounds on the tantalum tube ID surface were identified. They were TaC, Ta_2O_5 (β form) and $TaNi_3$. Other compounds were present in the tantalum tube ID coating, however, they could not be identified. Since the only major element on the tantalum ID aside from tantalum was Ni, some form of nickel-tantalum compound apparently made up the metallic layer confirming the previously described microprobe analysis. The Ta_2O_5 (β) present in the tantalum tube ID probably resulted from air contamination entering the system through the previously mentioned leaks which probably existed for some time without detection. The only potential source of carbon, found as TaC, was the possible contamination of the system by oil. Subsequent tantalum tube heating could have decomposed the oil, depositing a carbon-rich film on the surface which subsequently combined with the tantalum forming the compound TaC.

VII. EFFECT OF CHEMICAL CLEANING

The effectiveness of chemical cleaning performed on Test Section No. 4 and 5 should be viewed in terms, not only of the resultant subsequent heat transfer performance, but also in terms of the presumed system conditions during the run following the cleaning process. This is so because chemical cleaning could remove any heat transfer inhibiting surface film, however, its effect would be completely nullified if the system contained an air leak, or allowed oil back diffusion, during the subsequent restart. In a great many instances, the integrity of the loop during the restart, following cleaning of these test sections became questionable when a leak was detected in the system shortly after restart.

The chemical cleaning procedure used on these test sections was originally developed for the removal of decomposed oil residues from 9Cr-1Mo steel, SNAP-8 boiler tube surfaces. It had proven effective several times in full scale boiler tests. Prior to its use on tantalum, laboratory tests confirmed the effectiveness of the procedure, not only for removal of oil but also for surface tantalum oxides which were rapidly formed above 400°F. As a result of the initial experience gained in cleaning and subsequent operation of the tantalum test sections, a deficiency in the procedure when applied to tantalum became apparent. This deficiency in cleaning procedures and subsequent operation of the test system was not apparent when used for 9Cr-1Mo steel. The cleaning procedure as initially developed used the first three chemicals shown in Section III-C. However, these were followed by a single final rinse with sodium hydroxide. This original procedure was used to clean Test Section No. 4 during fabrication. It appeared from the initial poor heat performance that the cleaning was not effective. After approximately 2 hours of operation an abrupt (within 15 minutes) improvement in performance occurred.

It was postulated at that time that the cleaning procedure actually left a film on the tantalum surface and that it required the 2 hour operating period for the flowing mercury to mechanically scrub this film off. Subsequent laboratory tests indicated that the sodium hydroxide formed a

sodium tantalate which is not readily soluble in water. Thus the subsequent final water flush did not thoroughly clean the tantalum surface. To correct this deficiency changes were made in this originally developed cleaning procedure leading to the ultimate procedure as described in Section III-C. First, potassium hydroxide was substituted for the sodium hydroxide. A potassium tantalate is formed but this compound is readily soluble in water. Second, additional alternating acid and hydroxide flushes were incorporated. The addition of neutralizing flushes insured the complete absence of any residual films.

Demonstration of the adequacy of the cleaning method was provided by the single chemical cleaning performed on Test Section No. 5. After 2693 hours of operation, including 10 restarts, which witnessed almost continuous degraded heat transfer performance, the test section was cleaned and the integrity of the mercury loop confirmed. Immediate rated heat transfer performance followed on restart. This performance continued unchanged for the remaining 165 hours of operation.

It was concluded that this cleaning procedure would remove contaminants from the tantalum tube wall which would otherwise inhibit heat transfer performance. The method appears desirable for cleaning during fabrication of boilers to insure predictable immediate performance upon start-up. It should be recognized, however, that this procedure removes film, and does not prevent their formation. There is no substitute for a properly fabricated and high integrity test loop which will keep contaminants out.

VIII. MERCURY CORROSION OF TANTALUM

Post-test evaluation of all test sections indicated that no mercury corrosion of the tantalum tube wall had occurred. The methods used were metallography and actual pre and post-test wall thickness measurements.

Test Section No. 4 was the only section in which a marked reduction of tantalum tube wall thickness was measured at the end of the test. This reduction occurred at the downstream end of the section. A maximum wall loss of 0.0048 inch was measured. This was the test section on which the air injection test series was conducted and the wall loss was attributed to a tantalum/air reaction producing a powdery, non-adherent, tantalum surface oxide. The effect of this reaction was previously described in Section VI-A-3. It is sufficient here to point out that the cause was not a tantalum/mercury reaction.

The tantalum tube of Test Section No. 5 showed no apparent evidence of mercury corrosion attack. An occasional surface pit was found during post-test metallurgical evaluation (see Figure 18) but this was attributed to a pre-existing as-fabricated tube condition. Tube surface defects have been found during receiving inspection of raw material. These pits have not been considered detrimental to test section performance because of their minimal depth. The post-test evaluation of this test section confirmed this evaluation.

The tantalum-10% W wire of Test Section No. 5, contained some indications of attack. It is presently postulated that these are not the result of mercury corrosion. The test section operated for varying periods with air leaks in the system. The leaks occurred at the mercury outlet end of the test section, which was the location of the tantalum-10% W wire attack. Thus, a potential cause of cracking is air contamination. Second, the cracks may have been present in the as-fabricated section, produced by the helical coiling of the wire inside the tube or some other presently unknown operation in the test section fabrication procedure. Finally, laboratory experiments indicate that the ductility of the tantalum-10% W wire is greatly reduced as a result of hydrogen absorption during the chemical cleaning procedure. This hydrogen imbrittlement in combination with operating stresses could have resulted in surface cracks giving the appearance of Hg corrosion.

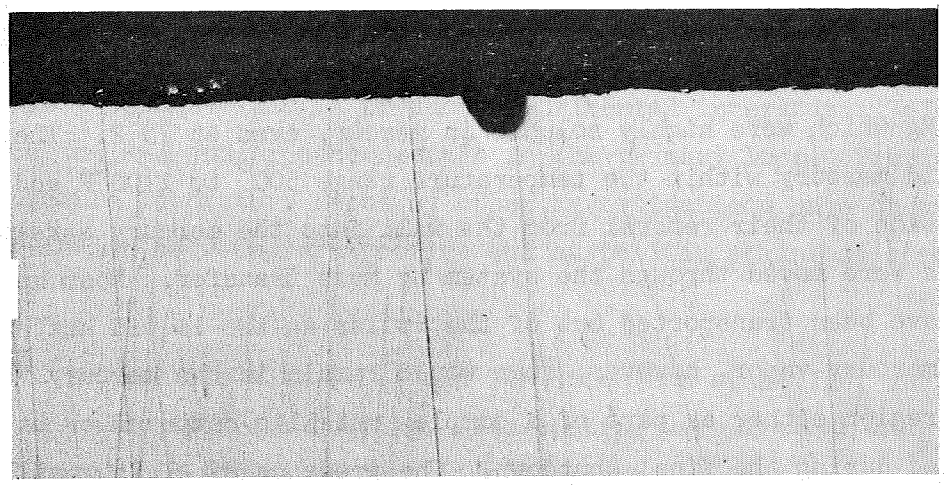


Figure 18. Hg Exposed Ta Tube Surface Pit in Test Section No. 5 After 2893 Hours Operation

The typical mercury corrosion reaction product of mass transfer which occurred in Test Section No. 5 was confirmed by the metallurgical evaluation of the surface films found on the tantalum tube I.D. The first four test sections were very short and did not contain a simulated tantalum lined mercury vapor superheater section. Therefore mass transfer deposits would not be anticipated.

The preoperational (as-received) metallic film on the tantalum tube I.D. surface of Test Section No. 5, shown in Figure 17, contained the elements Al and Mg which were highly soluble in mercury even at 75°F. They were exposed to liquid mercury within the temperature range 500° to 1100°F and there is no question of their removal from the wall into the mercury stream. The elements were moved through the system by mass transfer. Most material would have been transported out of the boiler in the liquid state entrained in the mercury vapor, however, some would remain in the mercury vapor superheater region either as part of a complex metallic compound or as residue deposited during the final shutdown. The presence of Al in small quantities was found throughout the test section in varying amounts of up to 0.1%. Mg could not be detected.

The Ni found on the tantalum tube wall in the superheater section of Test Section No. 5 was also postulated as a product of mercury mass transfer. The liquid leg piping of the mercury loop was fabricated from 316 stainless steel which contains a nominal 12% Ni in its composition. The recognized solubility of Ni in mercury is approximately 15 PPM at 500°F, the minimum liquid mercury temperature. Therefore, solution of this element into the mercury as it flows from the condenser to the boiler through the 316 stainless steel piping, could be anticipated. This Ni could come out of solution as the mercury vapor formed and deposit in this area of the test section. Apparently the subsequent reaction was Ni diffusion into the tantalum and formation of a nickel-tantalum compound. The compound should exhibit a hard brittle structure, typical of intermetallic compounds. However, this property could not be ascertained because of the thinness of the formed layer. In any event, there was no indication that this intermetallic layer was detrimental to continued test section operational integrity.

IX. OTHER MATERIALS EFFECTS-TEST SECTION NO. 5

A. STATIC NaK ENVELOPE

Test Section No. 5 was the sole representative of Basic Boiler Design Concept No. 2 (see Figure 3) which contained NaK in contact with the tantalum tube O.D. This non-flowing NaK inventory acted as a heat transfer medium between the tantalum tube and the 321 stainless steel tube. The latter tube protected the tantalum against contact with the flowing NaK of the primary loop thus avoiding a presumed detrimental interstitial element transfer from the NaK and/or other metals in the primary loop. Previous static NaK capsule laboratory tests by other investigations have shown acceptable minimum interaction between refractories and austenitic stainless steels.

1. Tube Surface Evaluation

The tantalum O.D., and 321 stainless steel I.D. surfaces contained a visually apparent coating. The coating was studied by various analytical procedures to determine elemental content and crystallographic structure.

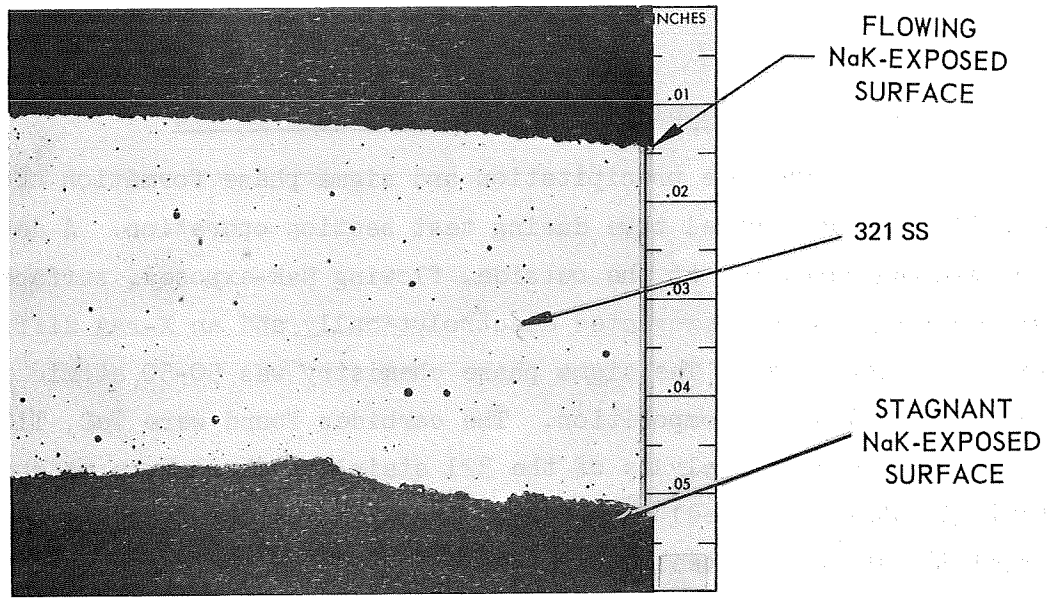
The major elements found on the tantalum tube O.D. surface were Ca, Ti, Fe and Mg, each between 1.1 and 2.2%. The elements present in trace amounts of less than 1% were Al, Nb, Ni, Zn and Cr. Electron diffraction analysis of the coating structure indicated the presence of only tantalum oxide, Ta_2O_5 (β form). However, studies by a second laboratory using X-ray diffraction techniques lead to the postulated theory that the coating on the tantalum O.D. was a residue left by diatomaceous earth or thermally decomposed asbestos. The presence of some foreign material of this nature was confirmed by detection of fibers in an electron micrograph of the residue. The implication of this latter theory was that during assembly of the test section a material similar to the thermal insulation wrapped around the section inadvertently entered the static NaK cavity. Subsequent 1300°F operation could have caused a reaction between the material and the tantalum with the NaK acting as the couplant.

It was suspected that the latter study was probably more valid, since the discovery in the initial study of Ta_2O_5 surface residue on the tantalum tube O.D. surface was not consistent with previous capsule tests. When tantalum was exposed to stagnant NaK, the NaK deoxidized the tantalum regardless of the relative oxygen content of the tantalum and NaK, thus negating the possibility of formation of a surface tantalum oxide.

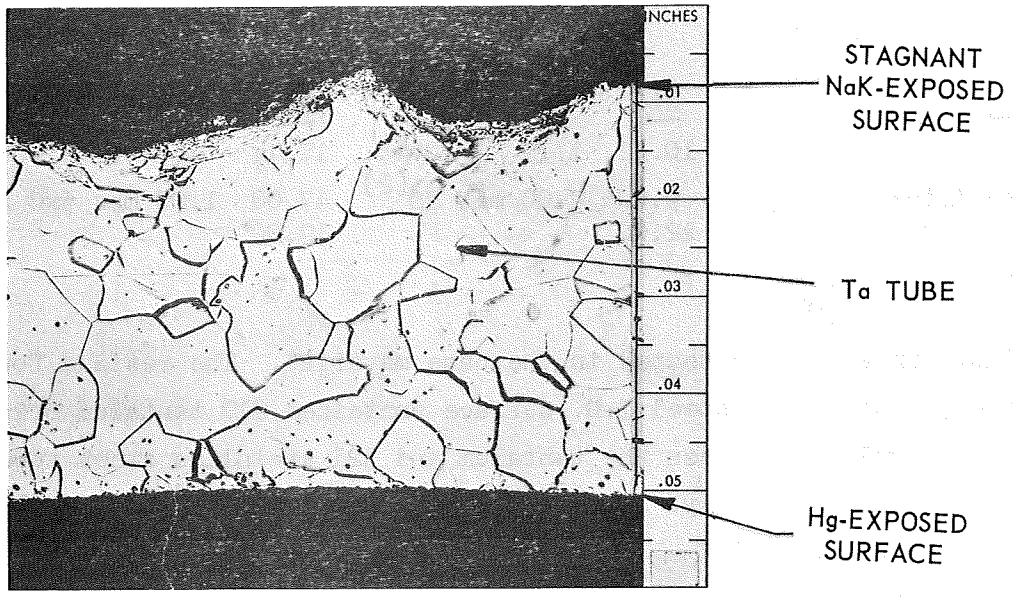
The 321 stainless steel oval tube I.D. surface contained a deposit identified by X-ray fluorescence and electron microprobe analysis as tantalum. Minor elements present were Fe, Cr, Ni, the constituent elements of 321 stainless steel. The X-ray diffraction analysis of 321 stainless steel tube I.D. surface residue indicated the presence of tantalum carbide. Tantalum carbide was not found on the O.D. surface of the tantalum tube, therefore, it was postulated that tantalum was transferred to the 321 stainless steel tube I.D. by galling of the two tubes during test section assembly. The tantalum is a strong carbide forming element and it was presumed that the carbide formed by interdiffusion with the free carbon in the 321 stainless steel.

Metallographic evaluation of the tube surfaces indicating areas of metal removal confirmed this theory of mechanical transference of tantalum. A typical location was as shown in Figure 19 where both the tantalum tube O.D. and corresponding location on the 321 stainless steel I.D. were affected. The cold worked distorted grains associated with the defect on the tantalum tube surface were presumed to have resulted by galling between the tube surfaces during test section assembly. Typical indications of 321 stainless steel tube galling were also visually observed without the benefit of magnification.

A final indication of galling between tubes was the bellows movement measurements taken during test section operation (see Section C, below).



S-451/24A TYPE 321 STAINLESS STEEL AS POLISHED



S-451/24B TANTALUM ETCHANT: 30% LACTIC
10% HF
10% HNO_3

Figure 19. Galled Opposing Ta and 321 Stainless Steel Surfaces in Test Section No. 5

2. 321 Stainless Steel Oval Tube Evaluation

Carbide precipitation and sigma phase formation occured in the 321 stainless steel tube during test section operation. A decarburized zone was developed at the outside, flowing Nak-exposed, surface. The sigma and carbides were extracted electrolytically and an X-ray diffraction analysis was conducted. The sigma phase chemistry was 50-50 atomic percent Fe-Cr, a typical sigma composition. The carbides found were TaC, TiC, $Cr_{23}C_6$. The chemical analysis of the 321 stainless steel tube matched the typical 321 stainless steel chemistry. Tantalum was not found in the as-received 321 stainless steel tubes (see below) which indicated that the tantalum carbides were probably the product of tantalum tube galling and carbon transport as discussed above.

Chemical Analysis of 321 Stainless Steel Oval Tube of Test Section No. 5

Sample Distance from Mercury Inlet (inches)	C	Ni	Mn	Cu	Cr	Si	Ti	Ta
100	.072	10.32	1.75	0.48	17.44	.50	.487	(1)
Typical Literature Analysis	.08 max	9.00 12.00	2.00 max	NS(2)	17.00 19.00	1.00 max	5xC Min	N.S.
As-received	.044 .047							N.D.(3)

(1) Tantalum was not detected in the base material. The residue found on the 321 stainless steel I.D. surface contained 61% tantalum originated in the galling between the tantalum and 321 stainless steel tube during Test Section assembly.

(2) N.S. - Not Specified

(3) N.D. - Not Detected

A moderate hardness increase apparently occurred during testing as shown below:

HARDNESS (ROCKWELL B) OF 321 SS OVAL TUBE IN TEST SECTION NO. 5

Sample Distance from Mercury Inlet (inches)	Hardness Location		
	I.D.	Center	O.D.
Typical literature value for annealed material	79	79	79
As-received	83	81	83
158	92	88	88
370	87	86	88

Tensile tests indicated the ultimate strength of the 321 stainless steel tube material was higher and ductility lower than typical literature values (see below).

75° TENSILE TESTS(1) ON UNWELDED 321 SS OVAL TUBE SPECIMENS REMOVED FROM TEST SECTION NO. 5

Sample Distance from Mercury Inlet (inches)	Yield Strength(2) (ksi)	Ultimate Strength (ksi)	Elongation (1% in 1 in)
53	50.8	93.3	25.8
141	47.5	91.7	21.0
282	46.6	91.5	18.4
298	46.4	79.8	12.4
357	46.4	93.3	47.2
Average of above 5 values	47.5	89.9	25.0
Typical /Literature data for Annealed Material	35	87	55

(1) Strain rate was .005 in./minute to yield and .05 in./min. thereafter to failure.

(2) Yield strength determined by drop-beam method.

The data indicated that the 2858 hrs. operating exposure at 1300°F of the 321 stainless steel oval tube had produced microstructural and mechanical

property changes which were typical and anticipated for this material. The precipitation of carbides and formation of sigma phase normally occur in austenitic stainless steels at this temperature. A consequential effect on mechanical properties, notably ductility reduction, results from these microstructural changes. There was no indication, however, that the decrease in ductility would continue, with further exposure to 40,000 hours, at a rate which represents a life limiting condition in a full size SNAP-8 boiler. Since present data was limited to 2858 hours of testing, additional tests should be conducted for reliable data extrapolation to predict operation to 10,000 hours or more.

B. TANTALUM/316 STAINLESS STEEL TRANSITION JOINT

The braze area (J-4800 cobalt base alloy, General Electric Co.) of both inlet and outlet transition joints were found defective after the test. However, comparison with an unexposed, as-fabricated, joint specimen, indicated that the deficiency probably existed prior to the test, based on the presence of similar defects. A separation existed at the braze-tantalum interface on the I.D. side of the tantalum groove (see Figure 20) and braze alloy removal was noted at the mercury exposed faces of the joint. A question remains relative to the separation. It may not have resulted from test section operation. At various times during laboratory testing the brittleness of the braze alloy (even in the as-brazed condition) has resulted in braze layer cracking during specimen preparation. One consequence of exposure during testing, based on comparison of microhardness measurements, indicated that the 2858 hours operation had caused significant hardening and consequential reduced ductility of the braze at the braze/tantalum interface (see Figure 21).

The evaluation results indicate that the braze transition joint requires extensive additional testing before it would be recommended for use in a full size boiler requiring 40,000 hr. life and 100 restart cycles. The low braze ductility in the as-fabricated condition plus the apparently lowered ductility of the braze area during elevated temperature, 1300°F exposure makes the joint susceptible to failure as a result of any operating transient which mechanically shocks the structure.

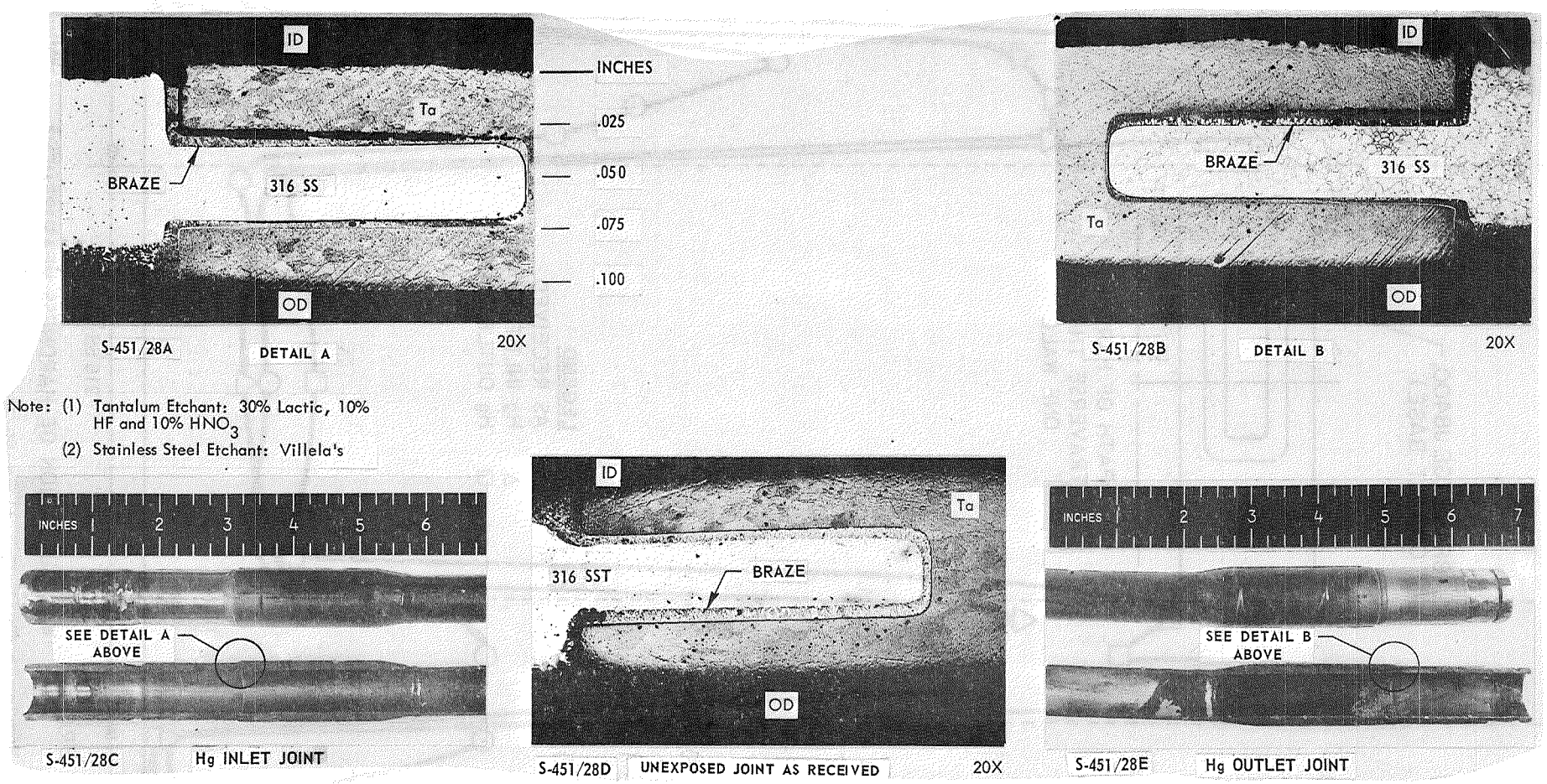


Figure 20. Inlet and Outlet Transition Joints of Test Section No. 5

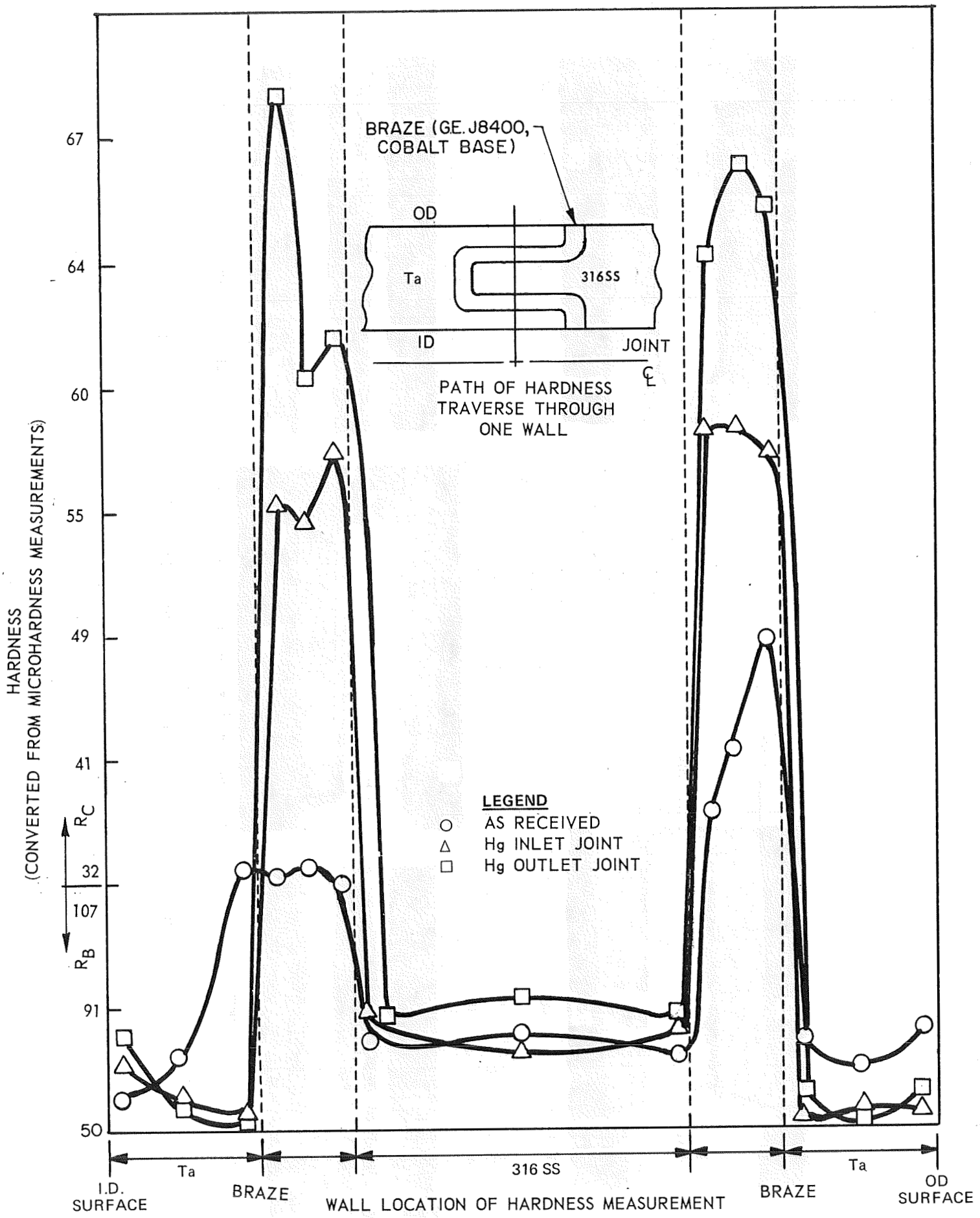


Figure 21. Hardness Traverse through Wall of Hg Inlet and Outlet Ta/316 Stainless Steel Tubular Transition Joints of Test Section No. 5

C. MERCURY INLET END BELLOWS

The thermal expansion compensator bellows was unaffected by operational exposure. The bellows was incorporated to compensate for the longitudinal differential expansion between the 316 stainless steel outer jacket/321 stainless steel oval tube unit structure and the tantalum mercury containment tube. It appeared adequate for the application based on this test.

X. FUTURE TEST PLANS

Several additional tests are planned or are in progress to further evaluate the mercury containment tube concepts of Test Sections No. 4 and 5. First, a full size single tube test in the Seventh Scale Loop facility is in process to further evaluate the explosive bonded tube concept of Test Section No. 4. Since the explosive bonding technique is presently insufficiently developed to produce the tube lengths required for SNAP-8 boiler use, this test section also includes a bimetal tube butt joint developed to connect shorter tube lengths. Second, a duplicate test of Test Section No. 5 is planned. Finally, three full size SNAP-8 boilers have been fabricated based on the design principle of Test Section No. 5. These boilers are currently operating successfully in various SNAP-8 test facilities. Complete destruction evaluation of all these boilers is planned after completion of the required tests.

XI. CONCLUSIONS

A test program was conducted at Aerojet-San Ramon Nuclear Operations to evaluate the use of a tantalum tube for mercury (Hg) containment in the SNAP-8 boiler. Five tests on subscale SNAP-8 simulated boiler tube configurations were conducted. Various candidate basic boiler tube design concepts, and the effects of potential mercury loop contaminants on tantalum were evaluated. A summary of the conclusions derived from the test program is presented below:

. Tantalum appears to be sufficiently resistant to mercury corrosion and erosion under SNAP-8 boiler operating conditions to provide at least 40,000 hours of service.

. A swaged, unbonded, bimetal tube of tantalum inside 316 stainless steel is not acceptable as a mercury containment tube for the SNAP-8 boiler because of mercury attack of the 316 stainless steel clad and poor heat transfer characteristics. Mercury attack results from bypass mercury flow between the tubes and poor heat transfer results when this bypass mercury flow is prevented.

. A bimetal tube consisting of a tantalum liner bonded by either explosive bonding or by hot coextrusion to the I.D. surface of a 316 stainless steel tube appears to be acceptable as a mercury containment tube for the SNAP-8 boiler.

. A bimetal tube bonded by hot coextrusion is preferred to one bonded by the explosive bonding method because the latter contains pre-existing unbonded areas, and a life-limiting phenomena of debonding may occur during boiler operation.

. Ultrasonic inspection is an adequate tool for locating unbonded areas in bonded bimetal tubes fabricated by hot coextrusion or explosive bonding. Post-test evaluation using ultrasonic inspection data relating to unbonded areas in bimetal tubes could be unreliable if tantalum contamination occurred during operation which produced extensive tantalum tube cracking.

. A mercury containment tube assembly consisting of two non-concentric tubes of tantalum inside 321 stainless steel, with a stagnant NaK inventory filling an annular space between the tubes is acceptable for use in a SNAP-8 boiler requiring at least 40,000 hour operating life.

. The fabrication technique for the non-concentric tantalum - 321 stainless steel multi-tube test section was not adequate to prevent galling of the tubes which produced localized tube wall thinning.

. The manufacturer of the tantalum tubing used processing techniques which produced tube surface defects. Although no failure resulted, more stringent control of tube quality appears necessary.

. A brazed tantalum to 316 stainless steel transition joint (using J-4800 cobalt based braze alloy, produced by General Electric Co.) appears to be questionalbe for use in the SNAP-8 boiler. Brazed joints contain an inherently brittle as-fabricated braze layer which increases in hardness as a result of 1300°F exposure. The effect of stress induced by the difference in thermal expansion characteristics of tantalum and stainless steel must be more thoroughly evaluated.

. Degradation of heat transfer performance is caused by some unknown critical quantity of mix-4P3E organic fluid or air in the mercury system which forms a thermal barrier on the mercury exposed surface of the tantalum tube.

. Chemical cleaning effectively removes thermal barrier surface films from a tantalum Hg containment tube previously exposed to Mix-4P3E or air and restores full heat transfer capability of the system. However, the cleaning process causes hydrogen embrittlement of the tantalum-10% W turbulator wire causing a severe reduction in ductility.