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EVALUATION OF TANTALUM/316 STAINLESS STEEL BIMETALLIC TUBING

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Prepared by
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16. Abstract

Three lots of Ta/316 SS bimetallic tubing were thoroughly non-destructively and destructively evaluated. Two of the lots were fabricated by co-extrusion to size and the third by explosive bonding to size. Although dimensional control of all three lots of tubing was excellent, the lot co-extruded over a mandrel was superior to the other two lots in all respects.

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FOREWORD

The research described in this report was conducted by the Aerojet General Corporation under NASA contract NAS 3-10601 in support of the SNAP-8 mercury boiler being developed by Aerojet for NASA. The NASA Project Manager was Phillip L. Stone, of the Lewis Research Center Space Power Systems Division. The report was originally issued as Westinghouse report WANL-PR-PPP-001.

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I. INTRODUCTION AND SUMMARY

This report describes the work accomplished on Contract NAS 3-10601, "Evaluation of Tantalum/316 Stainless Steel Bimetallic Tubing". The bimetal tubing will be used to fabricate a prototype tube-in-tube mercury boiler for the SNAP-8 nuclear electric power system[1]. The purpose of this investigation was to evaluate tantalum/316 SS bimetal tubing made by different manufacturing techniques. Two lots of tubing were fabricated by extrusion to size and were produced by the Nuclear Metal Division of Whittaker Corp. (Lot A) and by Metallonics Corp. (Lot B) using different extrusion techniques. A third lot of tubing,(Lot C) was produced by Aerojet General Corp. by explosive bonding to size.

The overall quality of the Ta/316 bimetal tubing made by coextrusion to final size was superior to that of the explosively bonded to size tubing. The nature of the explosive bonding process resulted in inherent unbonded areas in the as fabricated tubing which propagated during subsequent thermal cycling. Also the explosive bonded to size tubing contained a nearly continuous layer of varying thickness of a brittle intermetallic compound at the bimetal interface.

The coextruded to size tubing exhibited a sound bimetal bond and no intermetallic layer was observed at the Ta/316 interface by optical metallography.

Of the three lots of tubing, the Lot A made by extruding over a mandrel was superior in all respects. The Lot B tubing, made by the filled billet technique, was similar to the Lot A except for surface defects and straightness.

All three lots of tubing had excellent dimensional properties with respect to OD, ID, wall thickness and concentricity of the tantalum and 316 stainless steel components.

Thermal expansion measurements indicated that the heavy outer wall section of 316 SS controls the thermal expansion characteristics of the bimetal tubing.

Internal pressure burst and creep burst tests showed no difference in strength between the three lots of tubing. The strength values obtained indicated that published values for stainless steel can be used to predict the performance of bimetal tubes.

II. BACKGROUND

Improved corrosion resistance was required on the mercury side of the SNAP-8 NaK heated tube-in-tube mercury boiler[1]. Since the refractory metals, columbium and tantalum, exhibited excellent corrosion resistance to boiling mercury and the austenitic stainless steels were resistant to 1350°F (732°C) NaK, a sound bimetal combination of the two materials in tubular form promised a solution to the corrosion problem.

As the development work proceeded, metallurgically bonded bimetal tubing was produced by hot extrusion and cold drawing to size and an investigation at WANL[2] had indicated that columbium/316 SS bimetal tubes could be weld joined in the basic configurations required for the SNAP-8 boiler.

An evaluation at WANL[3] of the thermal stability of the bimetal bond of a wide range of refractory metal/heat resistant alloy combinations indicated that tantalum exhibited excellent stability with the stabilized austenitic stainless steels at temperatures up to 1400°F (760°C) which is 50-100°F higher than the peak operating temperature anticipated for the SNAP-8 boiler. A preliminary fabrication evaluation at WANL had also indicated that although hot extrusion and explosive bonding would produce an acceptable bimetal interface, subsequent cold drawing and intermediate annealing operations tended to weaken the bimetal bond.

Based in part on this prior work at WANL, Ta/316 SS was selected as the optimum bimetal combination. Explosive bonding-to-size and extrusion-to-size were selected as the fabrication techniques for producing Ta/316 bimetal tubing.

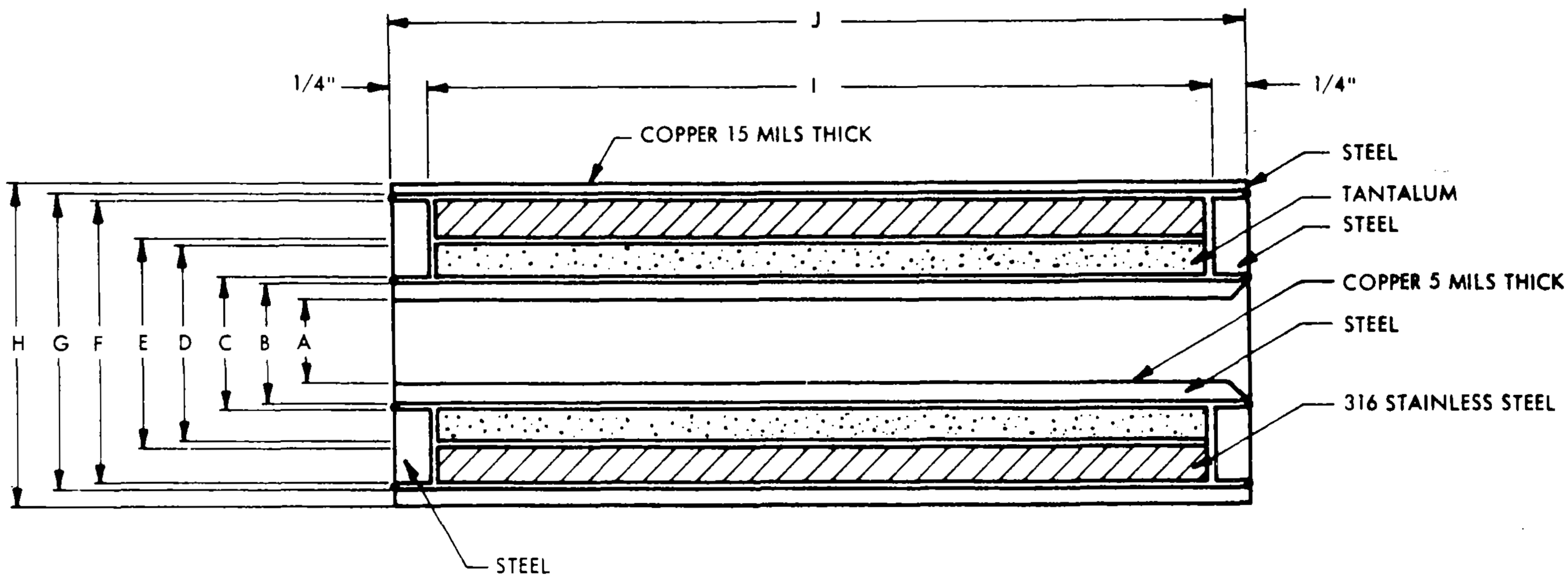
III. BIMETAL TUBING FABRICATION

The bimetal tubing evaluated was furnished by NASA. Approximately 200-300 feet of Lot A and Lot B tubing was produced in 15 foot lengths. The nominal tubing dimensions were 0.650 inch ID x 0.080 inch wall. The bimetal tube wall consisted of a 0.060 inch thick 316 stainless steel exterior and a 0.020 inch thick interior layer of unalloyed tantalum.

From Lot C, a single 9 foot length of explosively bonded tubing was received for evaluation. This length of tube was identified by the vendor as typical of the product. The nominal dimensions were 0.661 inch ID x 0.104 inch wall. The bimetal wall included a 0.020 inch interior tantalum layer and a 0.084 inch 316 stainless steel exterior layer.

Lot A. Lot A tubing fabricated by extruding to size over a mandrel, was produced by the Nuclear Metals Division of Whittaker Corporation, West Concord, Massachusetts. A copper canned hollow billet was heated to 1825°F (996°C) and extruded over a fixed mandrel. The billet design and extrusion details are shown in Figures 1 and 2. After outgassing at 800°F (427°C) in 10^{-4} torr vacuum, the billets were heated in a nitrogen atmosphere for 3-1/2 hours at 1825°F (996°C) prior to extrusion. Extrusion conditions are outlined in Table 1. After extrusion, the copper container was chemically removed using nitric acid and aqua regia. Stretcher straightening, chemical polishing of the tantalum, and centerless grinding of the stainless OD completed the fabrication process. The lot consisted of 13 tubes, each about 15 feet long. Chemical analyses for the starting materials are listed in Table 2.

Lot B. Lot B tubing, fabricated by filled billet extrusion, was produced by Metallonics Corp., Boston, Massachusetts. Basically a hollow bimetal billet was filled with mild steel, heated to 1850-1980°F (1010-1082°C), extruded and then the mild steel core was chemically removed. The extrusion billet design is shown in Figure 3 and the extrusion details are in



DIMENSIONS (INCHES)									
A	B	C	D	E	F	G	H	I	J
.657	1.063	1.073	1.662	1.670	2.750	2.760	2.970	8.00	8.50

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FIGURE 1 - Extrusion Billet Design Used For Lot A (Extruded over Mandrel) Tubing

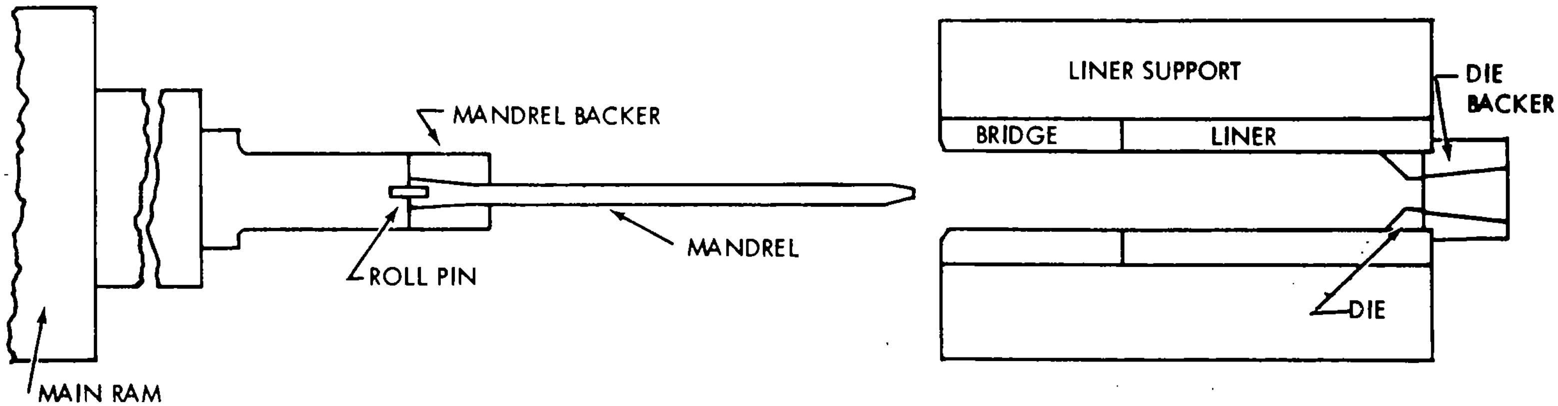


FIGURE 2 - Extrusion Set-Up Used for Producing the Lot A (Extruded Over Mandrel) Tubing

TABLE 1

EXTRUSION DATA FOR LOT A (EXTRUDED OVER MANDREL) TUBING

Tube No.	Extrusion Speed (ipm)	Extrusion Forces (Tons)	
		Upset	Run
14	400	600	560
20	550	550	675
21	600	675	600
22	500	725	650
23	500	700	650
24	500	700	650
25	500	675	650
26	500	725	675
27	500	700	650
28	500	700	650
29	500	700	650
30	500	700	650
31	500	725	650

Note - Other Extrusion Conditions
Extrusion Reduction - 25/1

Lubrication

Billet - sprayed dry film graphite
Liner - Necrolene
Die - Necrolene
Mandrel - Light coat of Lube-a-Tube

Liner ID - 3.050"
Mandrel Diameter - 0.630"
Die Size - 0. 857"

TABLE 2

CHEMICAL ANALYSIS FOR LOT A (EXTRUDED OVER MANDREL) RAW MATERIAL (VENDOR CERTIFICATIONS)

Tantalum, Lot 7106 (NRC)			
Analysis - ppm			
C	22	Cu	<1
O_2	28	Fe	11
N_2	26	Mo	<10
H_2	2	Nb	63
Al	<10	Ni	<1
Cr	<1	Si	<10
Ti	<5	W	170
316 Stainless Bar Heat No. 16240 (ARMCO)			
Analysis - weight percent			
C	0.055	Cr	16.96
Mn	1.60	Ni	13.24
P	0.029	Mo	2.74
S	0.021	Cu	0.31
Si	0.46	Co	0.19

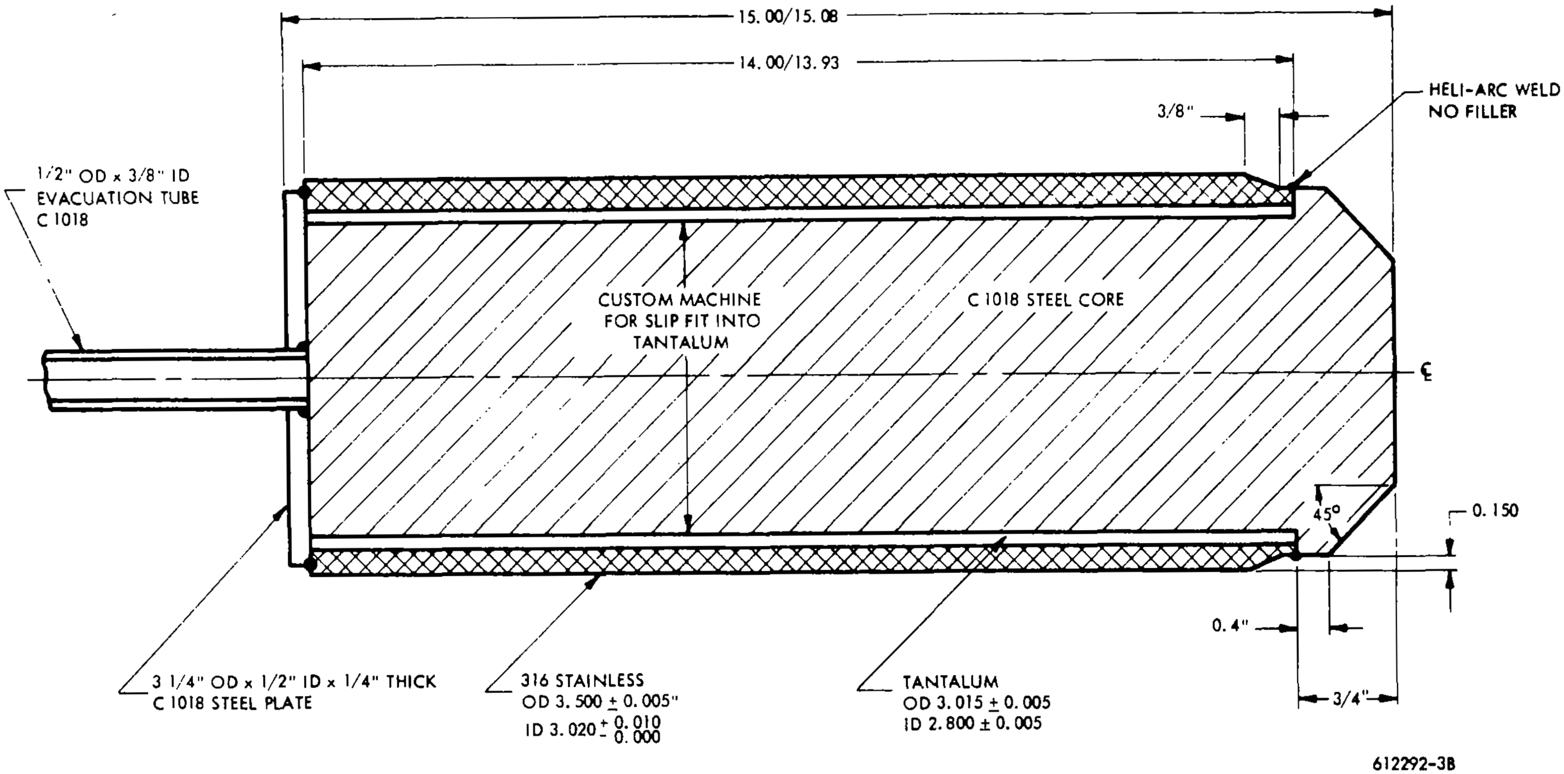


FIGURE 3 - Extrusion Billet Design For Lot B Filled Billet Extrusion

Table 3. The 316 stainless steel bimetal component of the tubing served as the protective outer shell of the billet. The exterior stainless steel cylinder was GTA welded to the mild steel core at the billet nose and to a 1/4 inch thick mild steel plate at the tail to seal the billet in preparation for evacuation. The billet was heated to 1000°F (538°C) during evacuation at 10^{-6} torr to provide outgassing prior to final sealing. The sealed billets were heated for 2-1/2 hours in an argon atmosphere at temperatures from 1850°F to 1980°F (1010-1082°C), (Table 3) and extruded through a zirconia die. The solid extrusions were straightened while hot and air cooled. The ends were cropped and the 1018 mild steel cores were dissolved with nitric acid at 200°F (93°C). The OD was sized by belt sanding and the ID by a torpedo mandrel. The ID surface was finished by alumina honing. The total lot consisted of 21 tubes, each about 15 feet long. Raw material chemical analyses are listed in Table 4.

Lot C. Lot C tubing, fabricated by explosively bonding to size, was produced by the Aerojet General Corp. at Downey, California. The explosive bonding process sequence is outlined in Figure 4 and detailed in Table 5. In preparation for explosive bonding a tantalum tube is placed inside an oversize stainless steel tube, producing a 0.045 inch annular spacing or "stand off" distance between the tubes. The uniform spacing is maintained by small protrusions spaced on the tantalum tube. The tantalum tube is filled with a low melting point alloy to provide support during forming and a layer of explosive charge is placed around the exterior 316 stainless steel tube. The protrusion or spacer areas do not bond because there is no stand off at these areas. The explosive charge is ignited from one end of the tube and bonding proceeds as shown below[5].

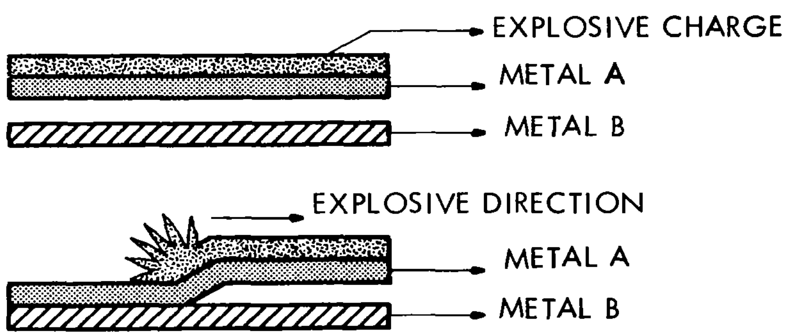


TABLE 3

EXTRUSION TEMPERATURE FOR LOT B (FILLED BILLET EXTRUSION) TUBING

Indicated Furnace Temperature*		Tube Number
1850°F	1010°C	2, X-12
1900°F	1038°C	M-4, M-5, M-7, M-9, X-13
1920°F	1049°C	M-15, M-16, M-17, M-18, M-19, M-20, M-21, M-22, M-6, M-10
1980°F	1082°C	M-3, M-11, M-12, M-13

* Temperatures listed are furnace temperatures and are 30-50°F higher than the actual billet temperature as determined by calibrated thermocouple readings.

Extrusion Ratio - 18/1

TABLE 4

RAW MATERIAL CHEMICAL ANALYSIS FOR LOT B (FILLED BILLET EXTRUSION) BIMETAL TUBING (VENDOR-FURNISHED)

Tantalum Tubing 3.015" OD x 2.80" ID Lot 6717			
Analysis - ppm			
C	25	O_2	<50
H_2	10	N_2	1.1
Cb	25	Ti	<5
Fe	<5	Mn	<5
Si	<5	Sn	N. D.
Ni	<5	Cr	<5
Ca	5	Na	N. D.
Al	<5	Mo	<5
Cu	<5	Zr	<5
Co	N. D.	Mg	<5
B	<5	W	<100

316 Pipe, 3" Sch 80 (3-1/2" OD x 0.300" Wall) Heat No. 2P1300 (USS)			
Analysis - weight percent			
C	0.065	Ni	13.77
Mn	1.62	Cr	16.97
S	0.019	Mo	2.21
Si	0.58		

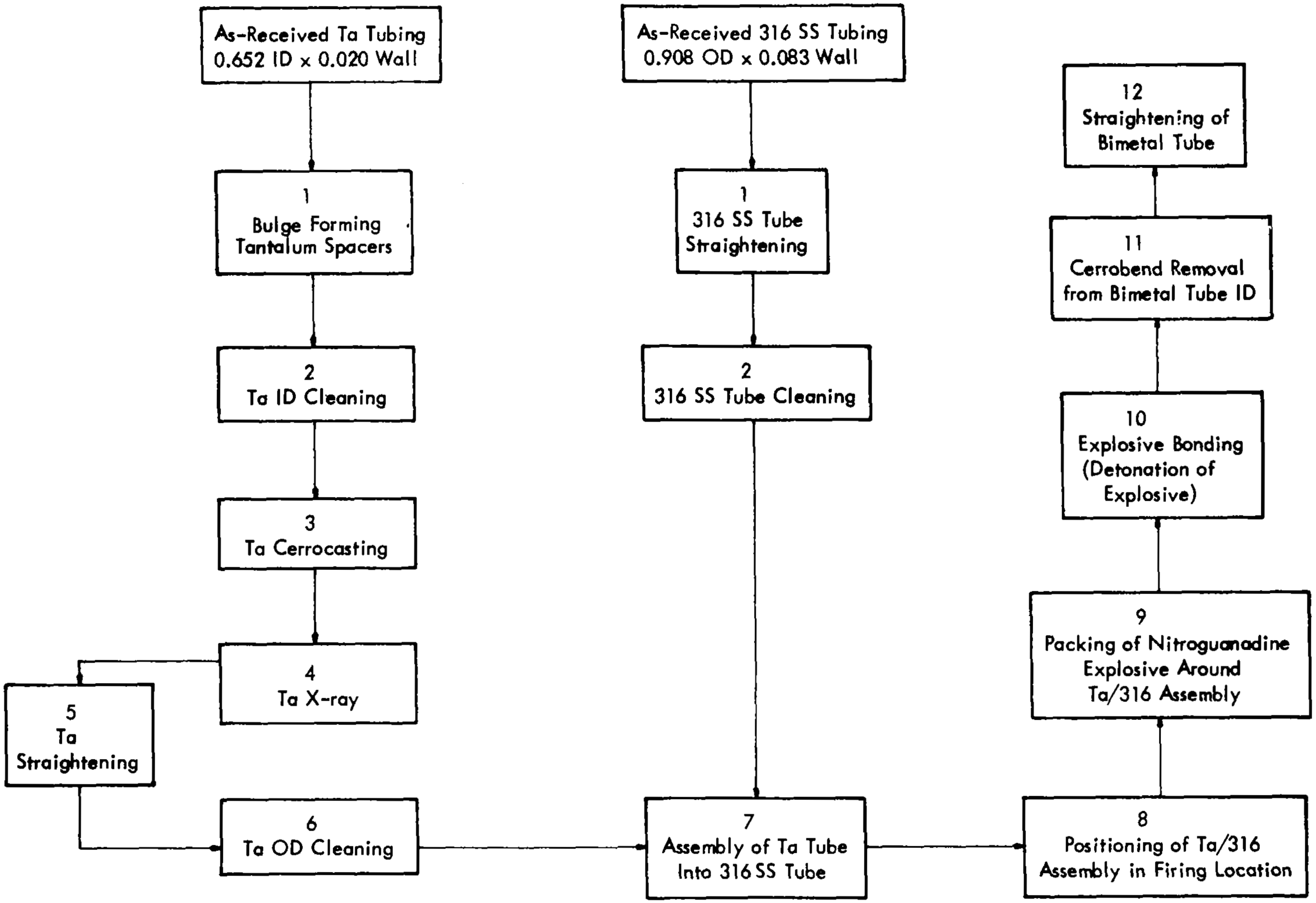


FIGURE 4 - Explosive Bonding Process for Lot C Tubing

TABLE 5

EXPLOSIVE BONDING OPERATION DETAILS

1. Bulging (introducing shallow, raised areas (<1/4" dia. on the tantalum tube OD) was accomplished by internal hydraulic pressurization of the tantalum tube causing selected localized expansion (.020 inch) of the Ta wall into a die cavity. Four dimples were formed with each dimpling operation to constitute one dimple pattern. The four dimples in each pattern are circumferentially distributed over a five inch length. The pattern is repetitive through the full tube length.

2. Each tube is then degreased.

3. Tantalum tubes placed in vertical position in a specially designed casting tower to fill ID with Cerrobend Alloy A*, to support Ta during the subsequent explosive bonding operation. Casting temperatures were monitored (thermocouples were spaced each 6 inches of tube length with temperature read-out on a multipoint recorder) to give cooling rate control by varying water flow in the cooling jacket of the casting tower.

4. X-ray inspection to ensure void-free cerrobend support material.
   X-ray exposure: 60 sec. at 100 kv
   Voids 0.010 inch rejectable
   Sanded and chemically etched

5. Tubes straightened to 0.005 inch/ft. TIR

6. The tantalum tube OD was chemically etched.

* Cerrobend composition: 50% Bi, 26.70% Pb, 13.30% Sn, 10% Cd
Cerrobend Melting Temperature = 158°F

TABLE 5 (CONTINUED)

EXPLOSIVE BONDING OPERATION DETAILS

7. Assembly performed vertically by lowering Ta tube into SS tube with protrusions centrally locating the Ta tube. Assembly was secured at ends using threaded fittings.

8. Ta/316 assembly positioned vertically in the center of a 3-1/2" dia. cardboard tube inside an oil drum assembly inside a firing tank 20 ft. dia. x 15 ft. deep.

9. Nitroguanadine powder explosive was packed around the Ta/316 assembly inside of the cardboard tube to a 7 gram per cubic inch density.

10. Explosive detonation and bonding.

11. Tube heated with hot water ($>158^{\circ}F$) to melt cerrobend.

One 9 foot tube representative of the fabrication process was received for evaluation.

IV. TEST PROGRAM

The test schedule followed for evaluating the bimetal tubing is outline 1 in Figure 5. Each lot of tubing was completely inspected non-destructively to determine bond integrity, surface condition and dimensions. Selected lengths of tubing from each lot were subjected to extensive destructive inspection and testing. This allowed evaluation of cladding thickness and uniformity, concentricity of bimetal layers, and further inspection of the ID surfaces. Durability of the bond was evaluated by thermal cycling. This consisted of heating the specimen to 1350°F (732°C) holding for 1 hour at temperature, cooling to 600°F (316°C) in 30 seconds and reheating to 1350°F (732°C). One hundred cycles constituted each test. Specimens were tested in the as-received condition and also after having been thermally treated 300 hours at 1650°F (899°C) to produce a 0.2 mil interdiffusion zone between the tantalum and 316 SS. The short time, high temperature thermal treatment simulates the diffusion zone width expected after 10^6 hours at 1350°F (732°C).

Creep burst properties at 1350°F (732°C) and thermal expansion characteristic between room temperature 1400°F (760°C) were also determined.

V. EXPERIMENTAL TECHNIQUES AND RESULTS

A. DIMENSIONAL MEASUREMENTS

Detailed measurements were made of the tubing dimensions. Measurement of the inside diameter of the tubes was made with a "Diagauge" pneumatic valve micrometer. The two point inside micrometer head operated an air valve which produced a variation in air pressure (read remotely) as a function of tubing diameter. The micrometer pressure gauge is calibrated in the tubing dimension range with a ring standard. The apparatus was attached to a long boom which permitted measurement along the entire length of tubing.

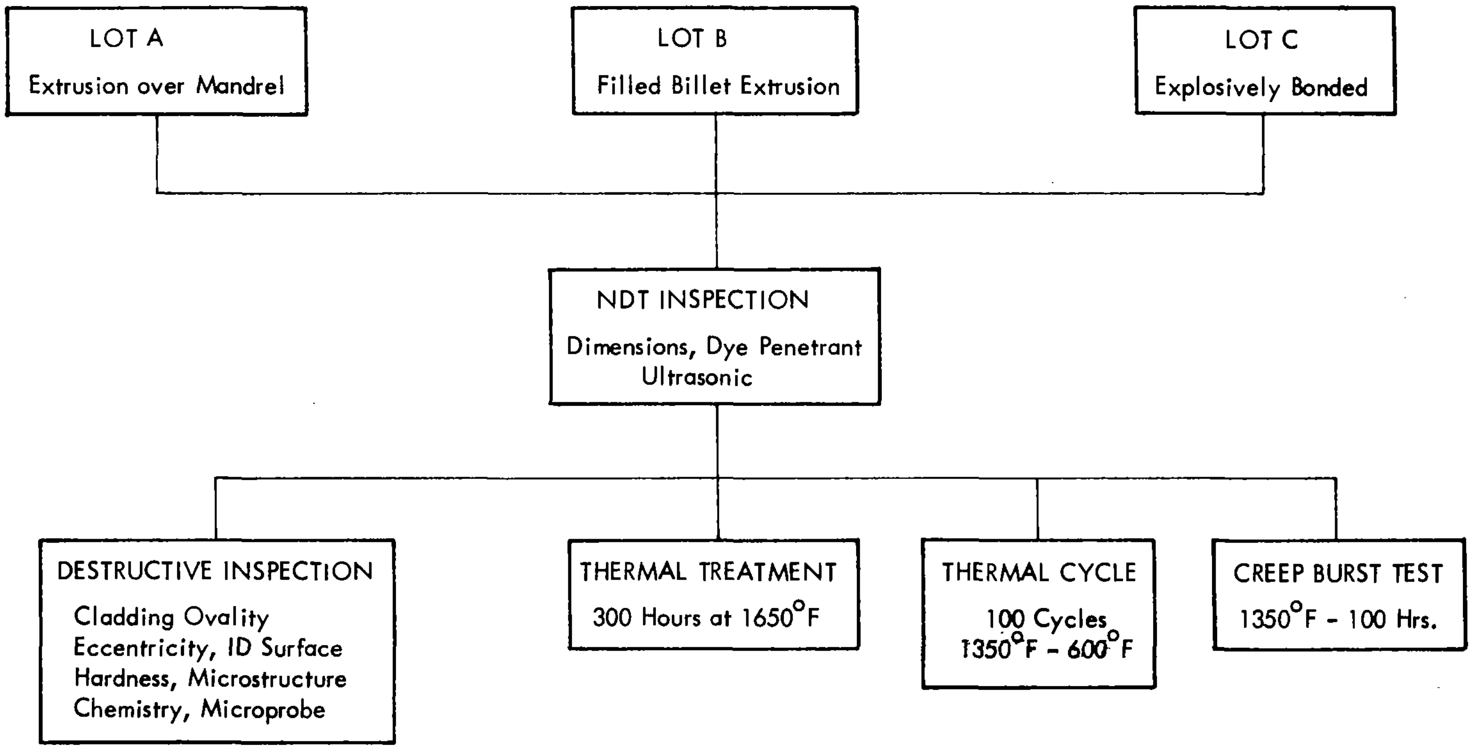


FIGURE 5 - Inspection and Test Schedule for Tantalum/316 Bimetal Stainless Steel Tubing

The entire Lot B and six of the tubes from Lot A required straightening to permit an accurate measurement of the inside diameter. Straightening of the deformed tubes did not introduce any defects.

The results of the dimensional measurements are shown in Table 6. All three types of tubing were produced with good dimensional control.

B. DYE-PENETRANT TESTING

Dye-penetrant inspection test results, for the Lot B tubing, are summarized in Table 7. The Lot A tubing and the Lot C tubing were essentially free of OD dye-penetrant indications. The test on Lot B however indicated numerous surface defects on about half of the tubes.

C. ULTRASONIC TESTING

Ultrasonic testing was used to determine the bond integrity between the stainless steel and refractory metal components. Differences in surface condition and interface geometry among the three types of tubing required the use of two ultrasonic test procedures in order to inspect for debonding at the bimetal interface. A "pulse-echo" technique satisfactorily delineated unbonded areas in the exploded-to-size tubing. However, the irregular interface found in both types of extruded tubing, Lot A and B as well as the rough ID surface of the Lot B tubing required a "pulse-echo-thickness measurement" technique in order to detect unbonded areas at the bimetal interface. An illustration of both procedures is shown in Figure 6.

The fifteen foot lengths of tubing were inspected in a semi-automatic fashion in a specially designed piece of equipment shown in Figure 7. The tubes were immersed in water and scanned using a focused transducer.

TABLE 6

DIMENSIONAL TEST RESULTS

LOT A (EXTRUDED OVER MANDREL) TUBING — 1

Tube No.	ID ave	σ ID	ID max/ave	ID min/ave	OD ave	σ OD	OD max/ave	OD min/ave	As-Received Length
	ID Measurements				OD Measurements				
14	.6447	.00029	.6461	.6433	.8001	.00039	.8028	.7995	16' - 4-3/4"
20	.6443	.00039	.6456	.6429	.8192	.00041	.8202	.8181	14' - 1-1/2"
21	.6424	.00048	.6435	.6412	.8164	.00075	.8177	.8180	14' - 4-1/2"
22	.6454	.00034	.6464	.6443	.8219	.00032	.8230	.8207	14' - 6-3/4"
23	.6449	.00029	.6458	.6440	.8168	.00042	.8177	.8159	14' - 7-3/4"
24	.6435	.00081	.6444	.6425	.8146	.00050	.8158	.8133	14' - 4-1/4"
25	.6428	.00041	.6441	.6414	.8161	.00043	.8170	.8151	14' - 9"
26	.6451	.00035	.6459	.6442	.8209	.00049	.8218	.8199	14' - 1-1/2"
27	.6472	.00051	.6484	.6459	.8231	.00076	.8248	.8214	14' - 5"(a)
28	.6440	.00028	.6450	.6430	.8124	.00053	.8155	.8128	14' - 7-1/2"
29	.6445	.00031	.6453	.6437	.8180	.00039	.8189	.8171	14' - 7-1/2"
30	.6452	.00035	.6464	.6440	.8204	.00051	.8223	.8184	14' - 4-1/2"
31	.6454	.00023	.6460	.6448	.8141	.00048	.8150	.8132	14' - 2-3/8"

(a) 6" cropped for dye penetrant check

LOT B (FILLED BILLET EXTRUSION) TUBING — 2

Tube No.	ID ave	σ ID	ID max/ave	ID min/ave	OD ave	σ OD	OD max/ave	OD min/ave	As-Received Length
	ID Measurements				OD Measurements				
M-3	.6552	.0004	.6566	.6537	.8188	.0003	.8197	.8180	15' - 9-1/2"(a)
M-4	.6526	.0002	.6532	.6508	.8177	.0004	.8193	.8162	14' - 10-1/2"
M-5	.6517	.0004	.6535	.6499	.8131	.0003	.8137	.8125	15' - 10-3/4"
M-6	.6533	.0004	.6552	.6514	.8194	.0002	.8200	.8187	15' - 9"
M-7	.6517	.0003	.6534	.6501	.8145	.0002	.8151	.8138	15' - 8-1/2"
M-9	.6524	.0006	.6545	.6503	.8158	.0009	.8168	.8147	15' - 5"
M-10	.6516	.0003	.6531	.6501	.8187	.0001	.8190	.8183	15' - 9-3/4"
M-11	.6504	.0003	.6516	.6492	.8156	.0001	.8164	.8147	15' - 10-1/4"
M-12	.6548	.0005	.6568	.6528	.8196	.0004	.8209	.8184	15' - 5"
M-13	.6516	.0003	.6529	.6502	.8170	.0002	.8177	.8162	15' - 10-1/4"(b)
M-15	.6531	.0002	.6542	.6520	.8184	.0001	.8186	.8182	15' - 10-1/4"
M-16	.6532	.0004	.6553	.6512	.8187	.0003	.8189	.8183	15' - 10-1/4"
M-17	.6526	.0003	.6542	.6511	.819[illegible]	.0004	.8197	.8186	15' - 5"
M-18	.6526	.0004	.6542	.6511	.8203	.0003	.8208	.8199	15' - 10-1/4"
M-19	.6530	.0007	.6550	.6509	.8184	.0002	.8189	.8178	15' - 10-1/4"
M-20	.6525	.0003	.6532	.6517	.8195	.0003	.8197	.8192	15' - 10-1/4"
M-21	.6535	.0006	.6564	.6506	.8211	.0002	.8220	.8201	15' - 10-1/4"
M-22	.6568	.0010	.6546	.6489	.8193	.0003	.8203	.8182	15' - 5-1/2"
X-12	.6530	.0010	.6556	.6504	.8143	.0010	.8145	.8140	16'
X-13	.6555	.0011	.6578	.6533	.7965	.0007	.7972	.7959	2'

(a) 11" cut out to remove unbond and OD crack
(b) 6-1/2" cut for dye penetrant check

LOT C (EXPLOSIVELY BONDED TUBING) — 3

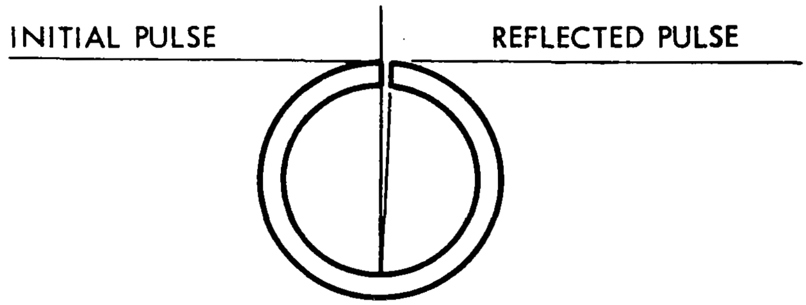
Tube No.	ID ave	σ ID	ID max/ave	ID min/ave	OD ave	σ OD	OD max/ave	OD min/ave	
	ID Measurements				OD Measurements				
96"	.6611	.0007	.6619	.6603	.8687	.0012	.8712	.8699	

TABLE 7

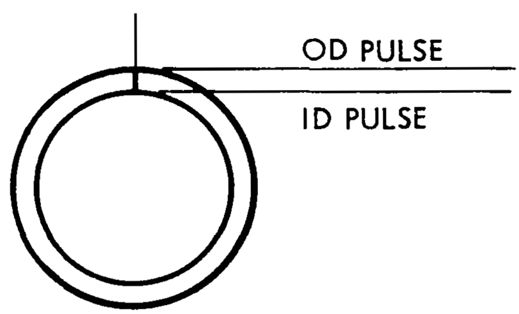
DYE PENETRANT RESULTS

Tube No.	LOT B (FILLED BILLET EXTRUSION) TUBING	
	Dye Penetrant	
	Defects[a]	Comments
M-3	32" longitudinal	all heavy indications
M-4	55" longitudinal 23" transverse	most are light indications
M-5	16" longitudinal	all heavy indications
M-6	11" longitudinal	all light indications
M-7	20" longitudinal	all light indications
M-9	41" longitudinal	some light some heavy indications
M-10	3" longitudinal	all light indications
M-11	32" longitudinal	all heavy indications
M-12	16" longitudinal 41" transverse	all light indications
M-13	40" longitudinal	most are light indications
M-15	23" longitudinal	all heavy indications
M-16	29" longitudinal	most are light indications
M-17	25" longitudinal	most are light indications
M-18	49" longitudinal	most are light indications
M-19	59" longitudinal	most are light indications
M-20	70" longitudinal	most are light indications
M-21	59" longitudinal	most are light indications
M-22	22" longitudinal	most are light indications
X-12	29" longitudinal	some are light; few heavy
X-13	7" longitudinal	all light indications
2	3" longitudinal	all light indications

(a) Numbers denote linear inches of longitudinal defects and transverse defects.



(a) SCHEMATIC OF PULSE ECHO TECHNIQUE USED TO ULTRASONICALLY INSPECT EXPLODED-TO-SIZE TUBING



(b) SCHEMATIC OF PULSE ECHO THICKNESS MEASUREMENT TECHNIQUE USED TO ULTRASONICALLY INSPECT EXTRUDED-TO-SIZE TUBING

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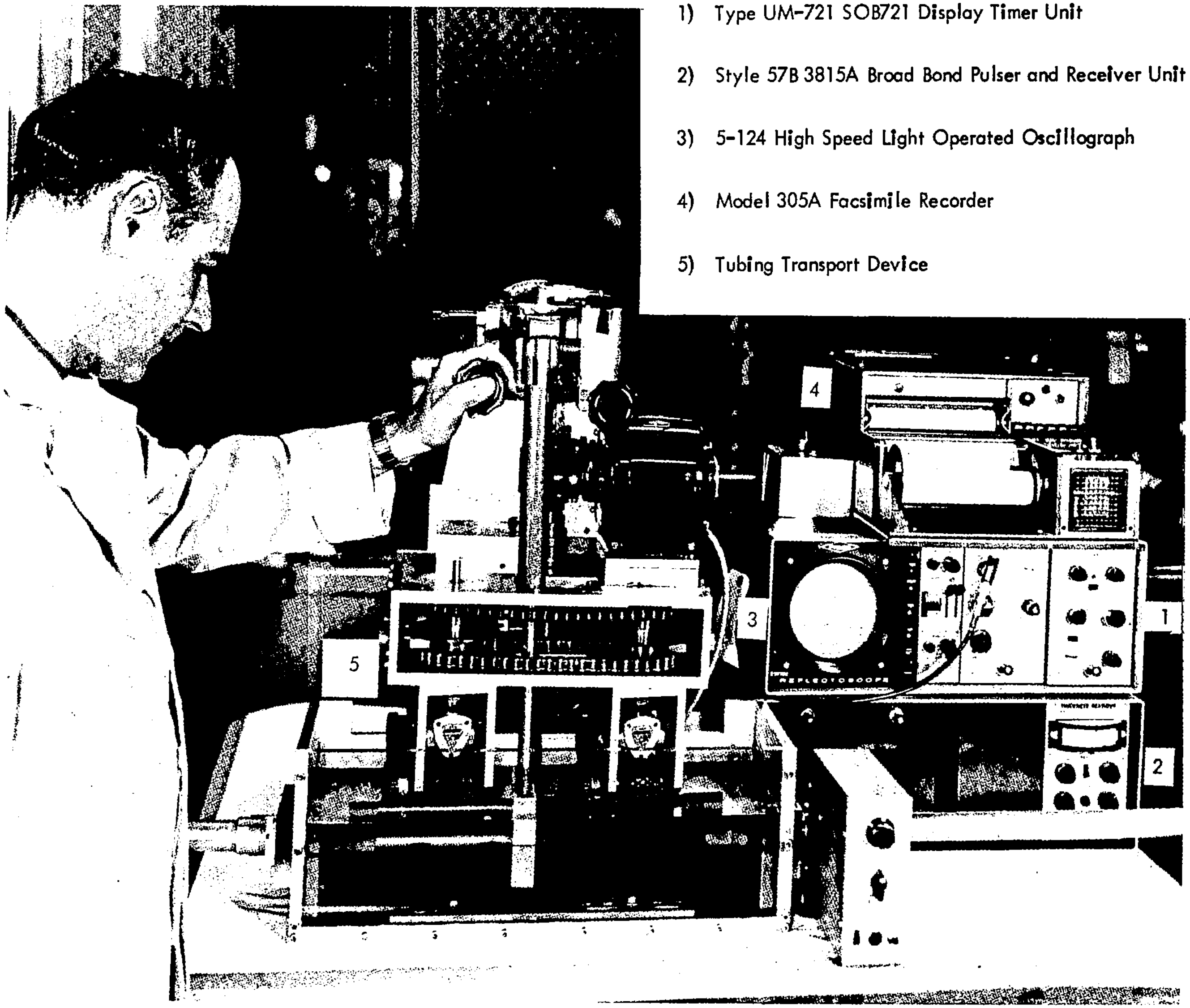
FIGURE 6 - Ultrasonic Inspection Techniques



1) Type UM-721 SOB721 Display Timer Unit

2) Style 57B 3815A Broad Bond Pulser and Receiver Unit

3) 5-124 High Speed Light Operated Oscillograph

4) Model 305A Facsimile Recorder

5) Tubing Transport Device

FIGURE 7 - Ultrasonic Test Apparatus

The tubes were fed into long slightly bowed booms, through seals into a water filled box. Here a rotating device moved the tubing past the test crystal in a helical fashion. The longitudinal speed and the pitch of the helix through the box could be varied from 0 ipm to 4 ipm and from 0"/360° to 1/2"/360° respectively. A spring loaded teflon vee block was used to maintain alignment between the crystal transducer and the tubing as well as fix the distance between the crystal and the tube.

The recording device used for both test techniques was a light operated oscilloscope, a high resolution galvanometer, and a strip chart recorder. It was assumed that any surface defects which occurred during extrusion and not completely removed during subsequent conditioning operations would interfere, thus a high resolution short pulser was necessary.

A Sperry Products Style 57A3621 crystal was used to inspect the Lot C (exploded to size) tubing and a Sperry Products Style SIL 57A2753 crystal and a Sperry Products Style 50E568 thickness read out module were used to inspect the co-extruded Lot A and Lot B tubing. Since no ultrasonic test standards were available it was necessary to manufacture a specimen from each type of tubing containing an unbond. This was necessary for two reasons: first to insure the differentiation between metallurgical bonding and intimate mechanical bonds; and second to fix the intensity of the pulses generated and the nature of the gating for the ID pulse. This gating was necessary to aid in distinguishing between varying degrees of bonding. The diameter of the test crystal was about 1/8 inch and so at any instant an area 1/8 inch in diameter of tubing was under inspection. Unbonded areas 1/8 inch or larger could be positively identified because the "echo pulse" was completely attentuated. Unbonded areas smaller than 1/8 inch in diameter were examined by means of the "gating system". Recalling that, at any instant an area 1/8 inch in diameter is under inspection, if only part of this area is unbonded then only part of the echo pulse is attenuated The gating system serves to completely attenuate the echo pulse if its intensity falls below a given value, thus giving an unbond signal in the recording apparatus. It is important to note that despite the use of gating, the determination of unbonded areas smaller than 1/8 inch in diameter where "partial bonding exists" is quite difficult and may be considered as the limit in resolution of the test technique.

Previous experience at WANL[4] had shown that debonding at the interfaces in extruded and drawn refractory metal/stainless steel bimetal tubing generally occurred over a continuous longitudinal length rather than as isolated discontinuous defects. An exception was the unbonding produced by the spacing protrusions in exploded-to-size tubing.

Test standards for each lot of tubing were made by the following techniques:

1. Explosively-Bonded-to-Size (Lot C Tubing): An interdiffusion zone approximately 0.2 mils thick was developed in a 6 inch long specimen. The test standard was then thermal cycled 20 times between 1350°F (732°C) and 600°F (316°C). The thermal cycling technique is described in detail in Section V-G. The thermal cycling of the diffusion layer produced unbonded areas in the tubing which generally propogated from the spacing protrusion areas and served as an ultrasonic test standard. In addition to this manufactured standard, the 96 inch long piece had previously been ultrasonically tested by Automation Industries and a copy of the trace generated was sent to WANL. This trace showed the existence of an unbonded area which was corroborated by the WANL results. Both specimens were retained as ultrasonic test standards.

2. Extruded-to-Size (Lot A and B Tubing): The extruded tubing had superior bond integrity and unbond areas could not be produced using the thermal exposure-thermal cycling technique. A mechanical unbond was produced as follows:

(a) A piece 1/2 inch long was cut from a 6 inch bimetallic tube.
(b) The stainless layer was etched from the 1/2 inch piece.
(c) A 1/2 inch deep counterbore removed the tantalum layer from the 5-1/2 inch long piece.

(d) The 1/2 inch long tantalum cylinder produced in operation (b) was inserted into the counterbore of the 5-1/2 inch long specimen.

The resulting 5-1/2 inch long specimen had a 1/2 inch unbond area at one end of the tube.

Ultrasonic Test Results. The ultrasonic inspection results are summarized in Table 8. The presence of unbonded areas, as determined by ultrasonic testing, was corroborated by metallography. An example is shown in Figure 8.

The extruded-to-size tubing, Lot A and B, had bond integrity superior to that of the explosively bonded-to-size tubing, Lot C. The explosively bonded tubing has unavoidable unbond areas at the spacing protrusion areas. In addition to these built in unbond areas, an unbonded area was found near the end in the 9 foot tube of this lot. In contrast, only a few small unbonded areas were found in Lot B and none in the Lot A tubing.

D. METALLOGRAPHIC, CHEMICAL AND MICROPROBE ANALYSES OF AS-RECEIVED SPECIMENS

Extensive metallographic examinations were conducted to define the nature of the interface and microstructure as well as determine the uniformity of the bimetal layers. Room temperature hardness traverses were made on mounted and polished specimens using a standard Tukon hardness machine with a 100 gm load. The hardness traverse results are shown in Figure 9. The explosively bonded-to-size tubing exhibited the highest hardness values for the 316 stainless steel component and were expected. This hardening is caused by shock loading during explosive bonding and has been discussed elsewhere[3,5,6]. The hardness values and metallographic examination of the Lot A and B tubing confirm that the stainless steel component partially recrystallized during extrusion. As expected, the hardness traverse in the stainless steel component of the coextruded to size tubing was relatively flat. The sharp hardness increase in the 316 stainless steel near the interface in the Lot C tubing is caused by localized plastic flow during the explosive bonding process[3,5,6].

TABLE 8
ULTRASONIC TEST RESULTS

TYPE B TUBING

Tube No.	Comments
M-3	Unbond at end 1/4" x 3/4", cut out
M-10	Unbond at 2 ft. from end 1/4" x 3/4"
X-13	Wall thickness fluctuations

All the other Metalonics tubes were fully bonded.(No U.T. indications)

TYPE C TUBING

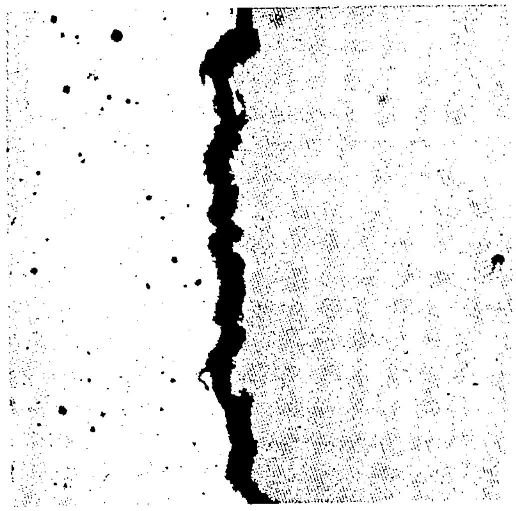
Tube No.	Comments
96"	Unbond formed at one end
43"	OK

All dimples in the exploded tubing indicated an area of unbound

TYPE A TUBING

No evidence of unbonded areas was found in any of the tubes.

316SS Tantalum



0.001" 400X

FIGURE 8 - Unbonded Area in Type B (Filled Billet Extrusion) Tubing Identified by Ultrasonic Testing.

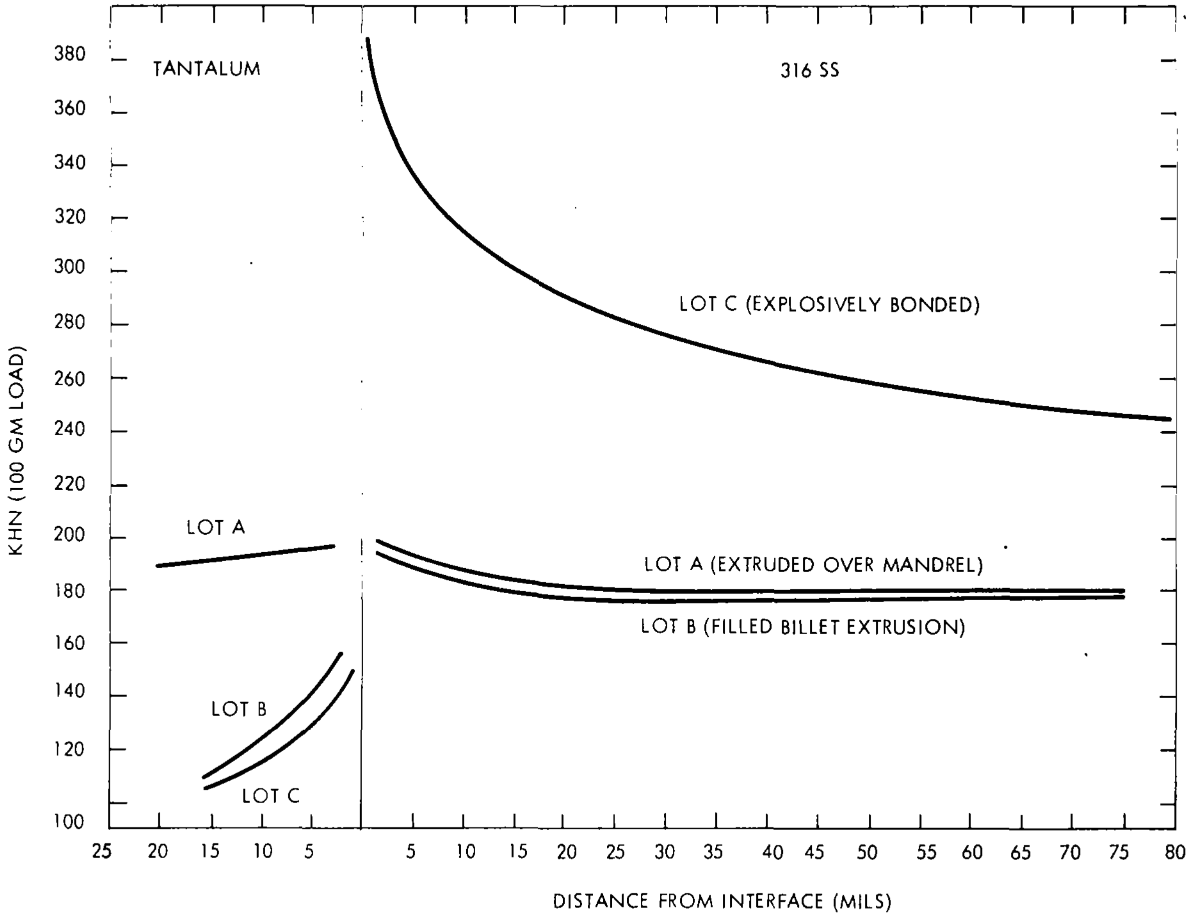


FIGURE 9 - Hardness Traverse of the As-Fabricated Tantalum/316SS Bimetal Layers

The high hardness values exhibited by the Lot A tantalum liner indicate that very little recovery and essentially no recrystallization occurred during extrusion; in contrast, the hardness level of the Lot B coextruded tubing is typical of partially recrystallized tantalum. The extrusion temperature of the Lot B tubing was 50 to 100F° higher than that of Lot A. This increment may account for the difference in the as-extruded hardness. The hardness gradient exhibited by the Lot C tantalum component was normal and this gradient is caused by localized plastic flow at the interface as discussed earlier. The hardness gradient in the Lot B tantalum liner may be caused by interstitial diffusion (C, O and N) from the stainless steel during heating for extrusion. The chemical analyses of the starting and as-fabricated carbon, oxygen and nitrogen levels are in Table 9. The as-fabricated Lot A and B tubing show only minor changes in the interstitial level. Although no starting analyses were available for the Lot C tubing, the very nature of the process would preclude significant chemistry changes during fabrication.

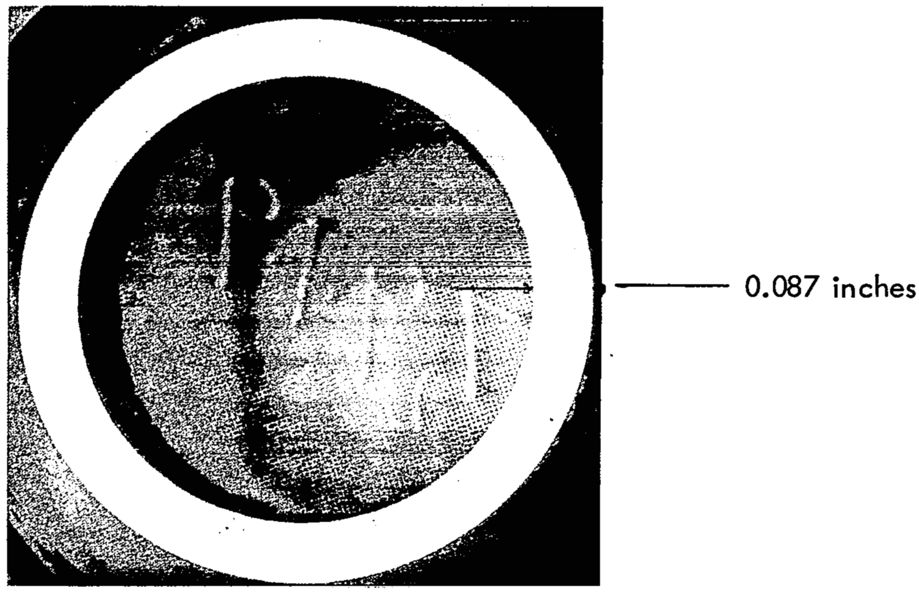
Lot A - Extruded Over Mandrel Tubing. The ID and OD surfaces were smooth although the bimetal interface was slightly irregular as shown in Figure 10. No evidence of ID or OD cracks, unbonded areas, or non-uniform bimetal layer thicknesses was found. Although the ID surfaces were smooth and free of cracks, irregularities probably caused by the metal flow characteristics during extrusion were observed (Figure 11a) and is typical of the ID surface. The 316 SS was partially recrystallized and fine grained and the tantalum was in the stress-relieved condition as shown in Figure 11b and 11c.

As shown in Table 9, there was no significant interstitial pickup by the tantalum component during fabrication. Electron microprobe traverse results did not reveal any detectable interdiffusion of the tantalum/316 stainless steel.

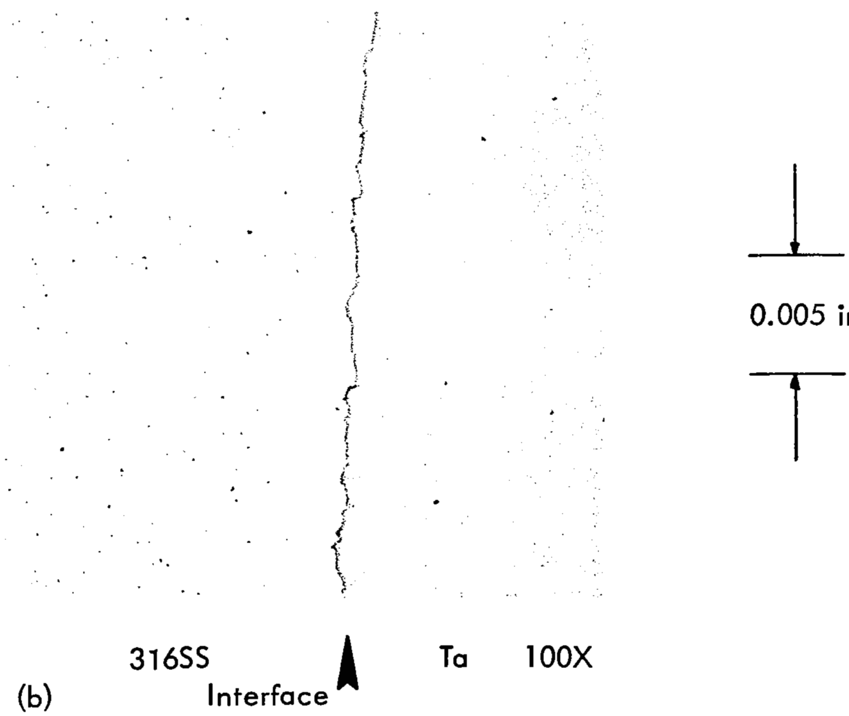
TABLE 9

CHEMICAL ANALYSES OF RAW MATERIAL AND FABRICATED BIMETAL TUBING

Specimen	Element	Analyses in Weight ppm		
		Type A Extruded Over Mandrel	Type B Filled Billet Extrusion	Type C Explosively Bonded
Tantalum Raw Material	C	22	25	--
	O	28	50	--
	N	26	1	--
Tantalum Fabricated Tubing	C	39	44	22
	O	53	69	77
	N	28	18	45
316SS Raw Material	C	550	650	---
	O	---	---	---
	N	---	---	---
316SS Fabricated Tubing	C	700	550	640
	O	70	120	140
	N	380	400	370

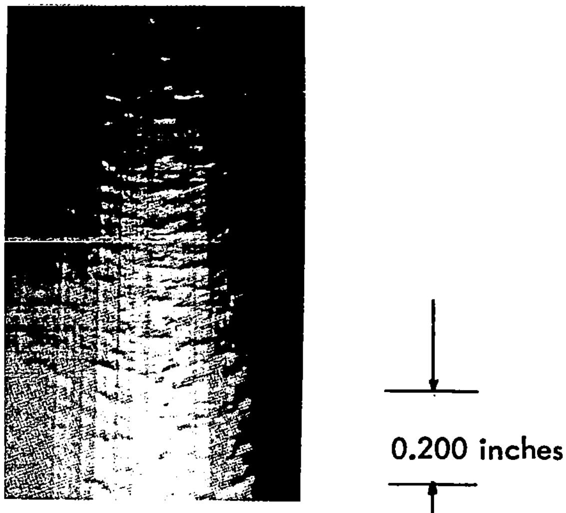


(a) Transverse Section

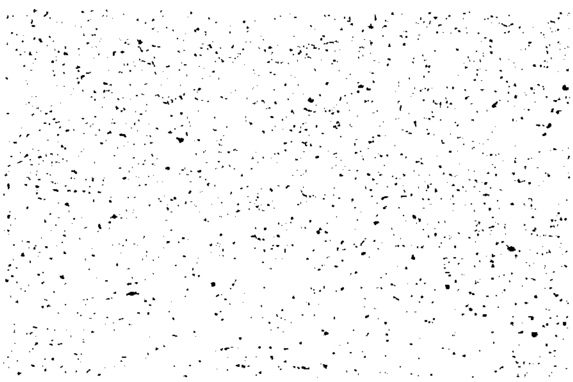


(b)

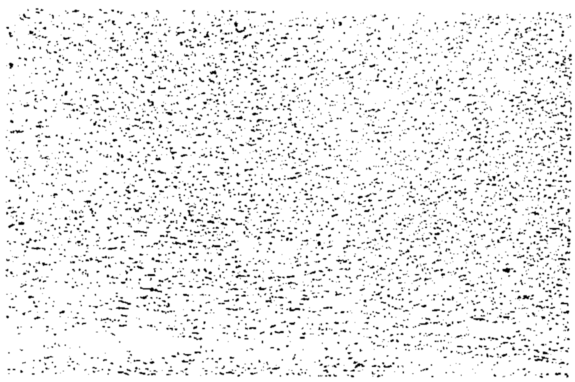
FIGURE 10 - Transverse Section of Lot A (Extruded Over Mandrel) Bimetal Tubing



(a) ID Surface



(b) 316SS Microstructure 100X



(c) Ta Microstructure 100X

FIGURE 11 - ID Surface and Microstructure of Type A (Extruded Over Mandrel) Ta/316SS Bimetal Tubing.

Lot B (Filled Billet Extrusion). The ID surface was rough and was formed of striations 2-3 mils deep as shown in Figures 12a and 13a. The OD surface was smooth. Both the OD and ID surfaces contained fissures of varying depths as shown in Figures 12b and 12c. The majority of ID cracks were about 2-3 mils deep. The 316 SS and Ta were fine grained and recrystallized as shown in Figure 13b and 13c.

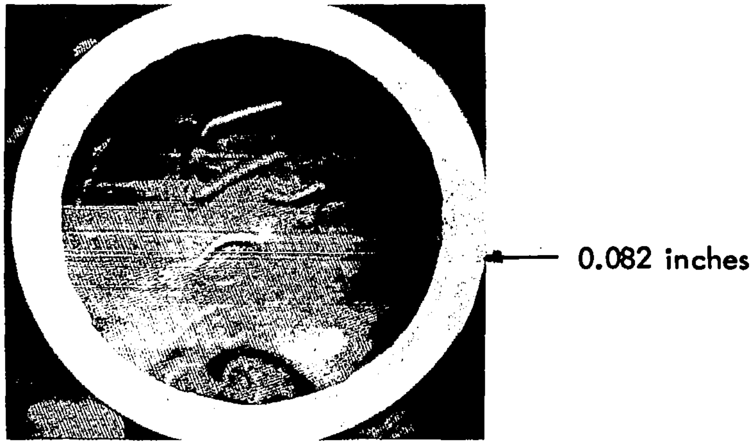
Chemical analysis did not indicate significant C, O, and N contamination of the Ta during fabrication as shown in Table 9. Microprobe analysis indicated there was no interdiffusion of Ta, Fe, Ni and Cr.

Lot C (Explosively Bonded). All exterior surfaces of the tubing were smooth and crack-free. Unlike the extruded tubing Lots A and B, the transverse section of the bimetal interface is ripple free as shown in Figure 14c. The original spacing protrusions required for explosive bonding are retained in the bonded tubing as shown in Figure 14a. The protrusions are the only disturbance to the very uniform bimetal layers. An ID surface view of a spacing protrusion is shown in Figure 14b.

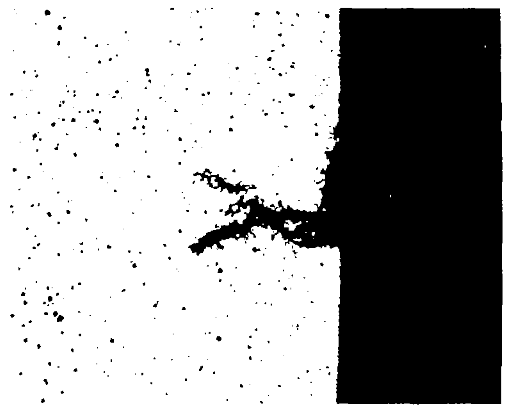
A nearly continuous intermetallic layer was observed at the bimetal interface of the as-explosively bonded tubing. Transverse cracks were observed in the intermetallic layer which occasionally propogaged into the tantalum as shown in Figure 15.

As shown in Figure 16, the tantalum and 316 SS components exhibit uniform equiaxed grains except near the interface which corresponds to the sharp increase in hardness shown in Figure 9.

Electron beam microprobe analyses showed that the interface layer (intermetallic compound) observed was a complex intermetallic compound containing Ta, Fe, Cr and Ni. Other than the intermetallic formation there was no interdiffusion of Ta, Fe, Ni and Cr detected.



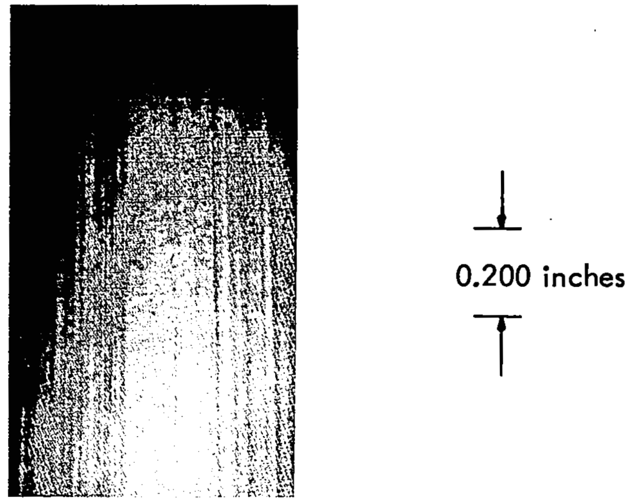
(a) Transverse Section



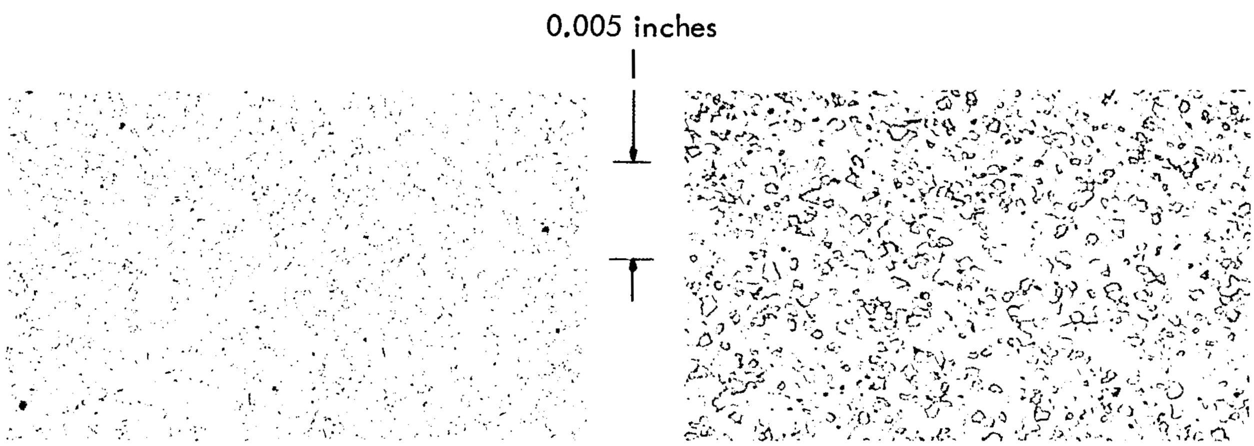
(b) ID Crack ~2 Mils Deep 100X

(c) OD Crack ~7-1/2 Mils Deep 100X

FIGURE 12 - Transverse Section of Lot B (Filled Billet Extrusion) Bimetal Tubing

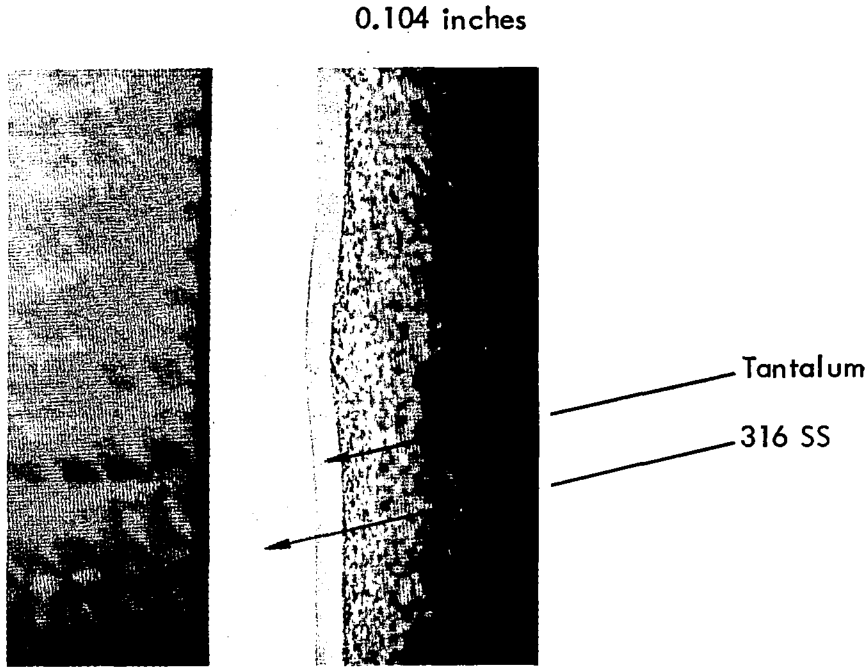


(a) ID Surface

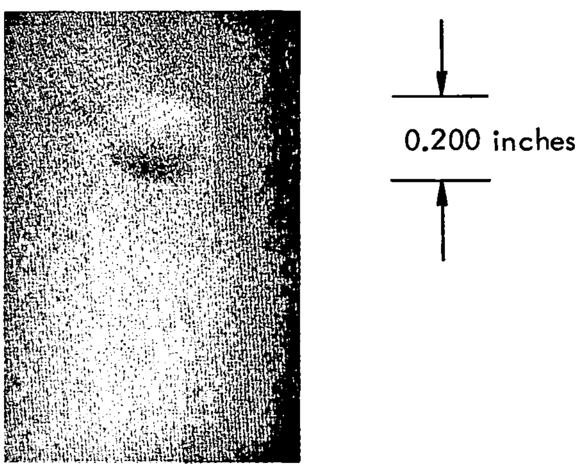


(b) 316 SS Microstructure 100X (c) Ta Microstructure 100X

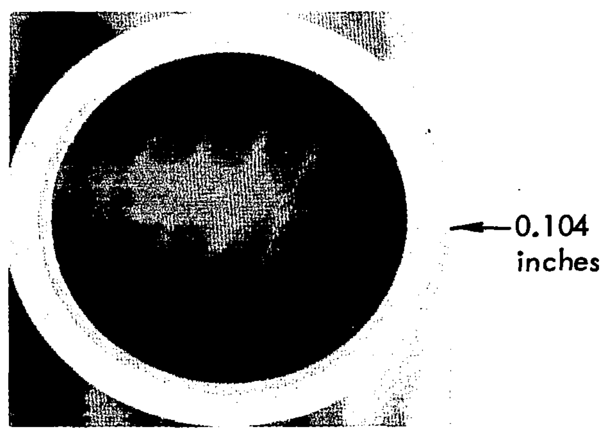
FIGURE 13 - ID Surface and Microstructure of Lot B (Filled Billet Extrusion) Bimetal Tubing



(a) Longitudinal Section Showing Spacer Protrusion



(b) ID Surface of Tantalum



(c)

FIGURE 14 - Sections of Lot C Explosively Bonded Bimetal Tubing

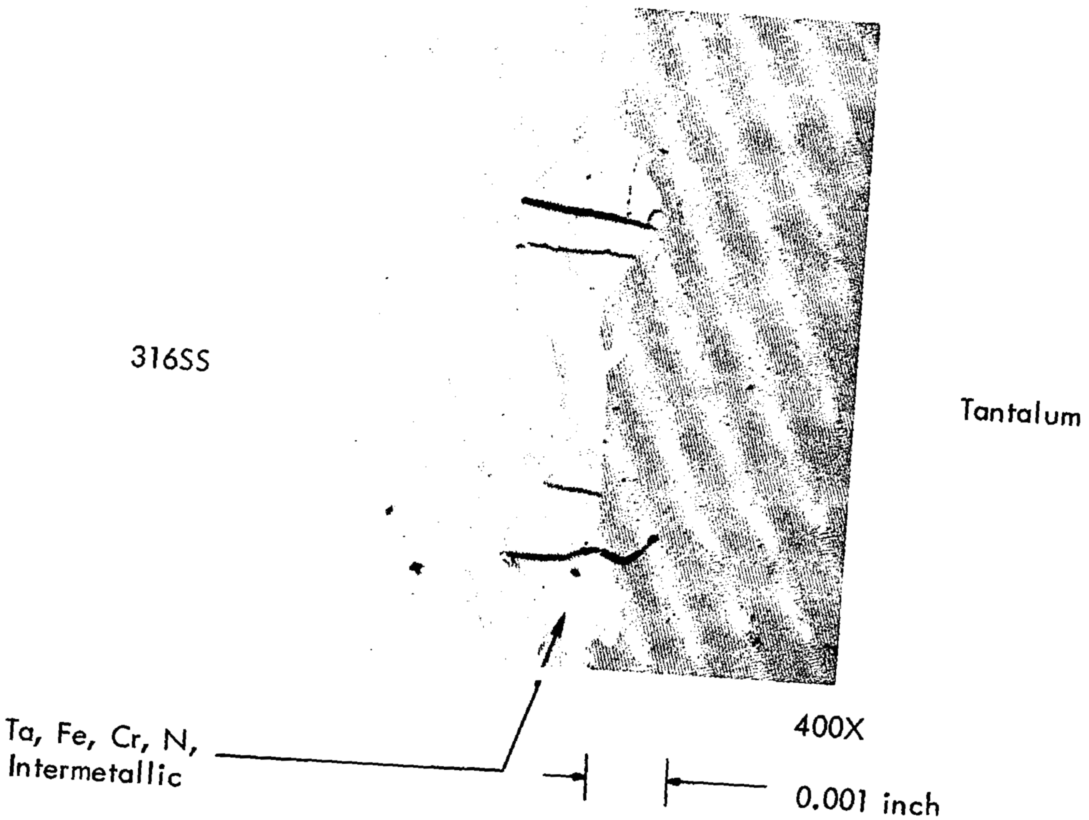
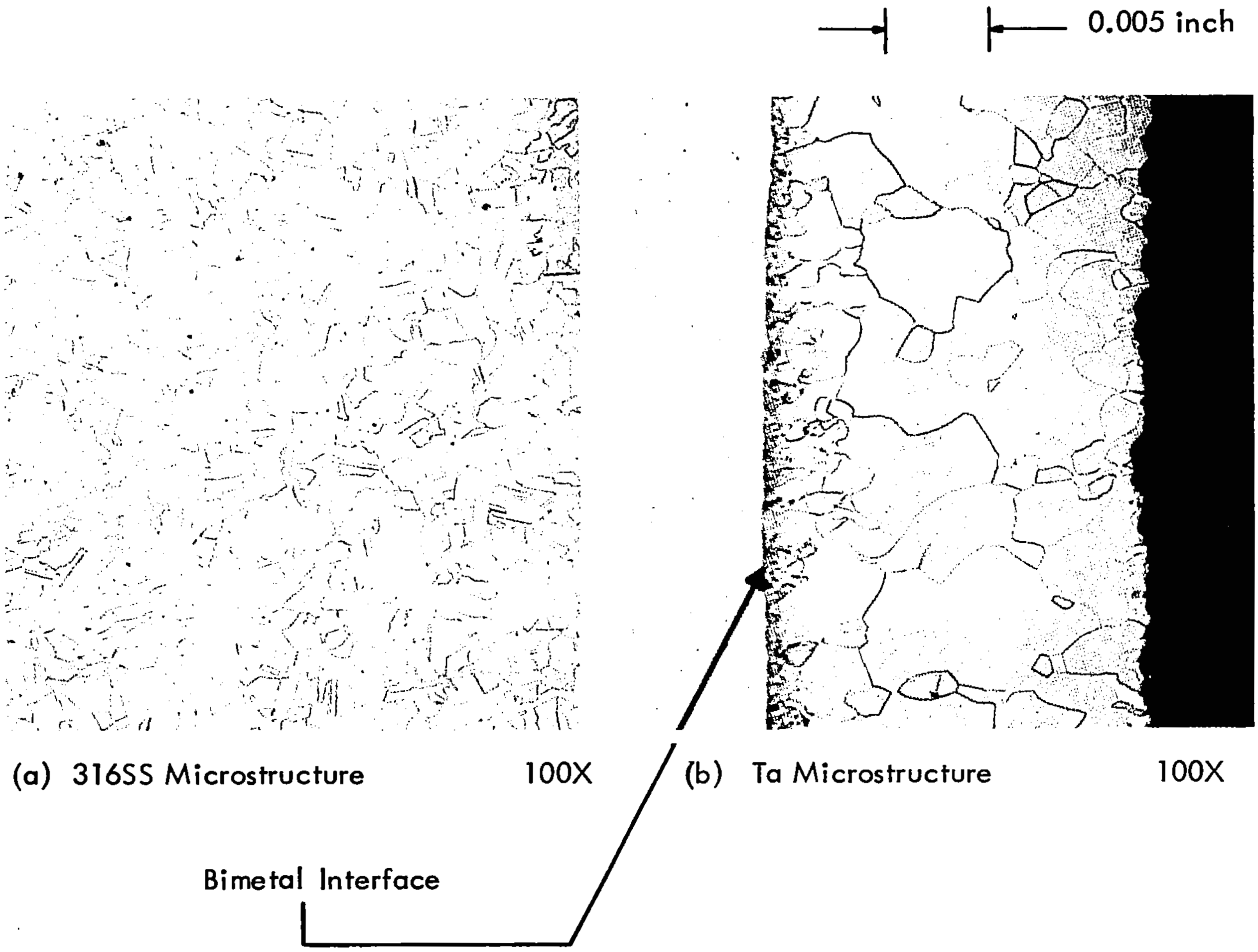


FIGURE 15 - Bimetal Interface in Type C Explosively Bonded Tubing



(a) 316SS Microstructure 100X

(b) Ta Microstructure 100X

FIGURE 16 - Microstructure of Type C Explosively Bonded Bimetal Tubing

E. THERMAL EXPANSION MEASUREMENTS

Thermal expansion determinations were made on the Ta/316 bimetal tubing in both the diametral and longitudinal direction. The thermal expansion measurement apparatus is shown in Figure 17. Thermal expansion measurements were made from room temperature to 1400°F (760°C). To prevent atmospheric contamination of refractory metal components, measurements were performed in a diffusion pumped vacuum system at a pressure of $\leq 1 \times 10^{-5}$ torr.

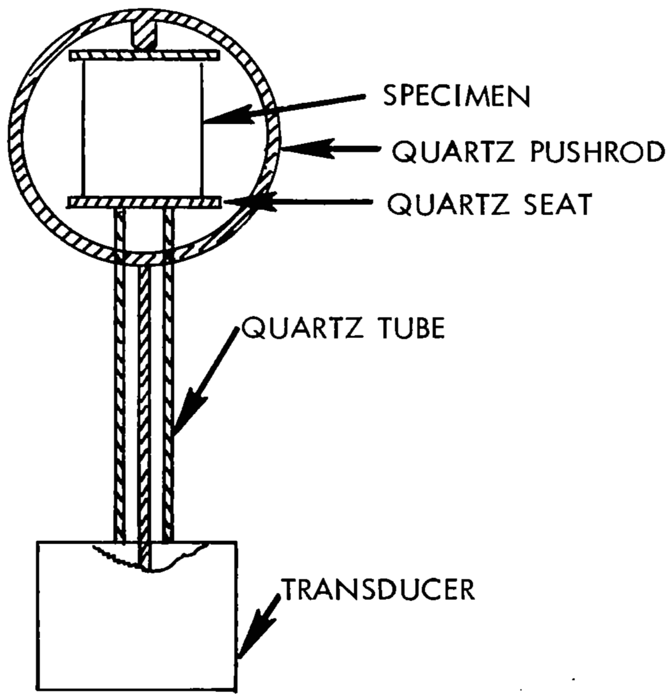
The unit utilizes a modified form of the standard quartz pushrod and tube technique. The quartz pushrod is formed into a ring to surround and contact the tubular specimen opposite the pedestal. The change in specimen diameter is thus transferred directly to pushrod motion. Pushrod motion is measured and recorded by an electro-mechanical transducer. The accuracy of the measurement is estimated to be $\pm 1\%$.

A vertical quartz tube with a flat polished upper end and a slit forms the pedestal. Figure 17a shows the apparatus with a bimetal specimen; Figure 17b shows schematically, the pushrod, tabs and transducer measuring a longitudinal specimen.

The thermal expansion results on the Lot A, mandrel extruded tubing, are shown in Figure 18. Repeated runs on the same specimen were comparable, indicating only a negligible effect from the preceding thermal exposure. The diametral expansion was also not significantly different considering that the tubing ovality could produce a greater variation in diametral thermal expansion measurements.

Similar measurements on Lot C, explosively bonded bimetal tubing, are shown in Figure 19. Again, no significant difference was observed between repeated runs on the same specimen or between diametral and longitudinal measurements.

Thermal Expansion Apparatus



Thermal Expansion Schematic

FIGURE 17 - Thermal Expansion Test Equipment

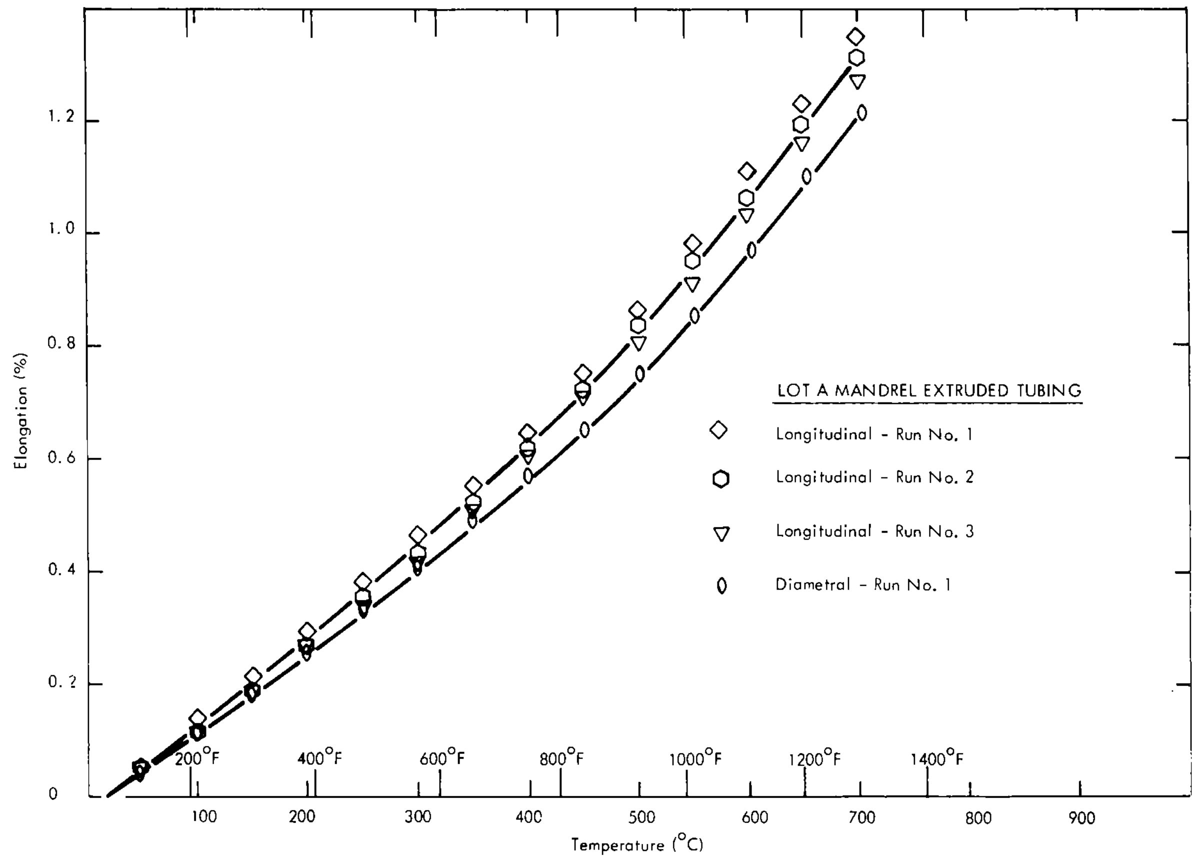


FIGURE 18 - Thermal Expansion Measurements of Lot A, Mandrel Extruded Tubing

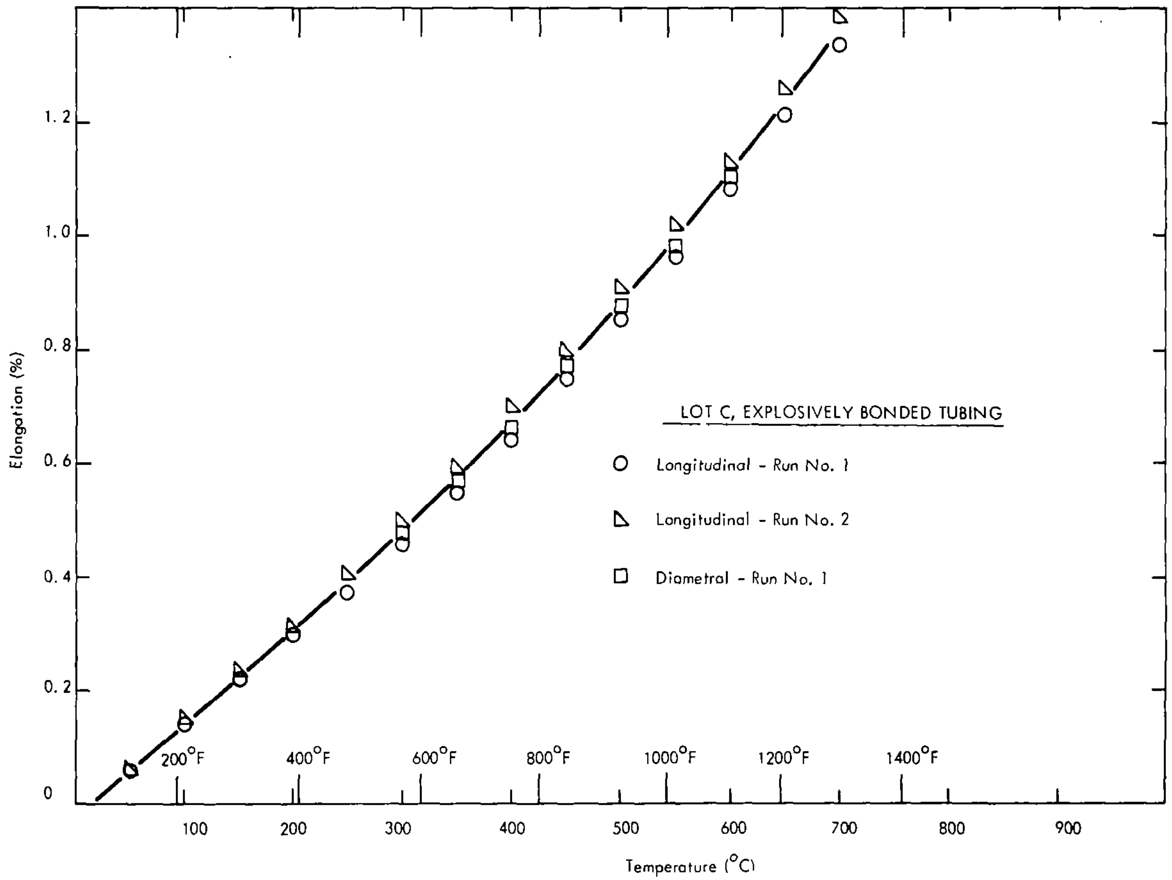


FIGURE 19 - Thermal Expansion Measurements of Type C, Explosively Bonded Tubing

The thermal expansion measurements of longitudinal specimens of Lot A, mandrel extruded, and Lot C, explosively bonded tubing, are compared in Figure 20. A section of 316 SS tubing prepared from Lot A bimetal tubing was run for comparison and it is evident that the thicker stainless steel section of the bimetal tubing dominates the bimetal thermal expansion characteristics. The average coefficient of thermal expansion measured for stainless steel from 100°F (38°C) to 1200°F (649°C) was 10.5 x 10^{-6} inches/inch/°F which agrees well with reference values of 10.3 x 10^{-6} inches/inch from 32°F to 1200°F (0°C to 649°C)[7]. For comparison, the thermal expansion of unalloyed tantalum over this temperature range is approximately one third that of 316 stainless steel or 3.6 x 10^{-6} inches/inch/°F.

F. PRESSURE TESTING

The apparatus used for creep-burst testing of the tubing is shown schematically in Figure 21. A detailed description of the equipment and operation can be found elsewhere[2]. Basically, the specimen, which is contained in an evacuated cylinder, is internally pressurized with high-purity helium gas and heated to the test temperature by using a hot-wall furnace. Several end plug designs were evaluated before the final design was fixed. The end plug employed was a 1/4 inch thick stainless steel disk. A counterbore 1/4 inch long was machined into the tantalum of each end of the specimen. The end plug was then placed into the resultant cavity and EB welded to the 316 SS layer. Short stainless steel support rings were slip-fit around each end to reinforce the welds. Figure 22 shows a schematic of the end plug design. Although a gas pressure seal was not made at the tantalum liner, there was no evidence that gas pressure ruptured the bimetal bond. The specimen size was chosen to produce a length to eliminate end effects on the diametral stress.

Two types of specimens are commonly used for pressure testing cylinders. As shown in Figure 23 the difference is in supported and unsupported end plugs. The unsupported specimen, as used exclusively in this program, adds a longitudinal stress component to the hoop stress. For relatively thin walled specimens, the longitudinal stress is one half the hoop stress. The resultant "effective" stress may be calculated by $\sigma_e = \sqrt{3}/2\ P\ r/t$[8] where

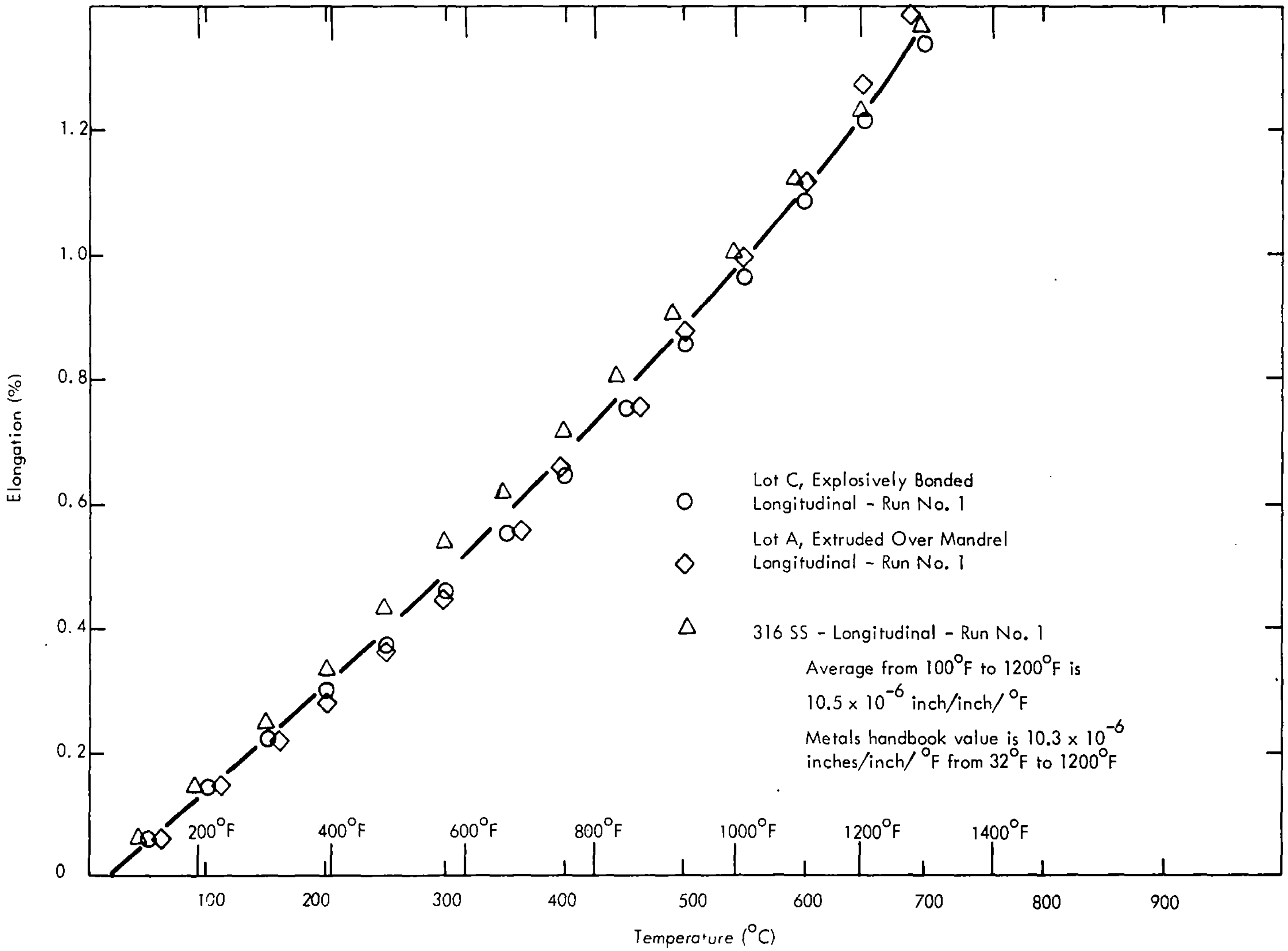


FIGURE 20 - Comparative Thermal Expansion Measurements of Bimetal Tubing

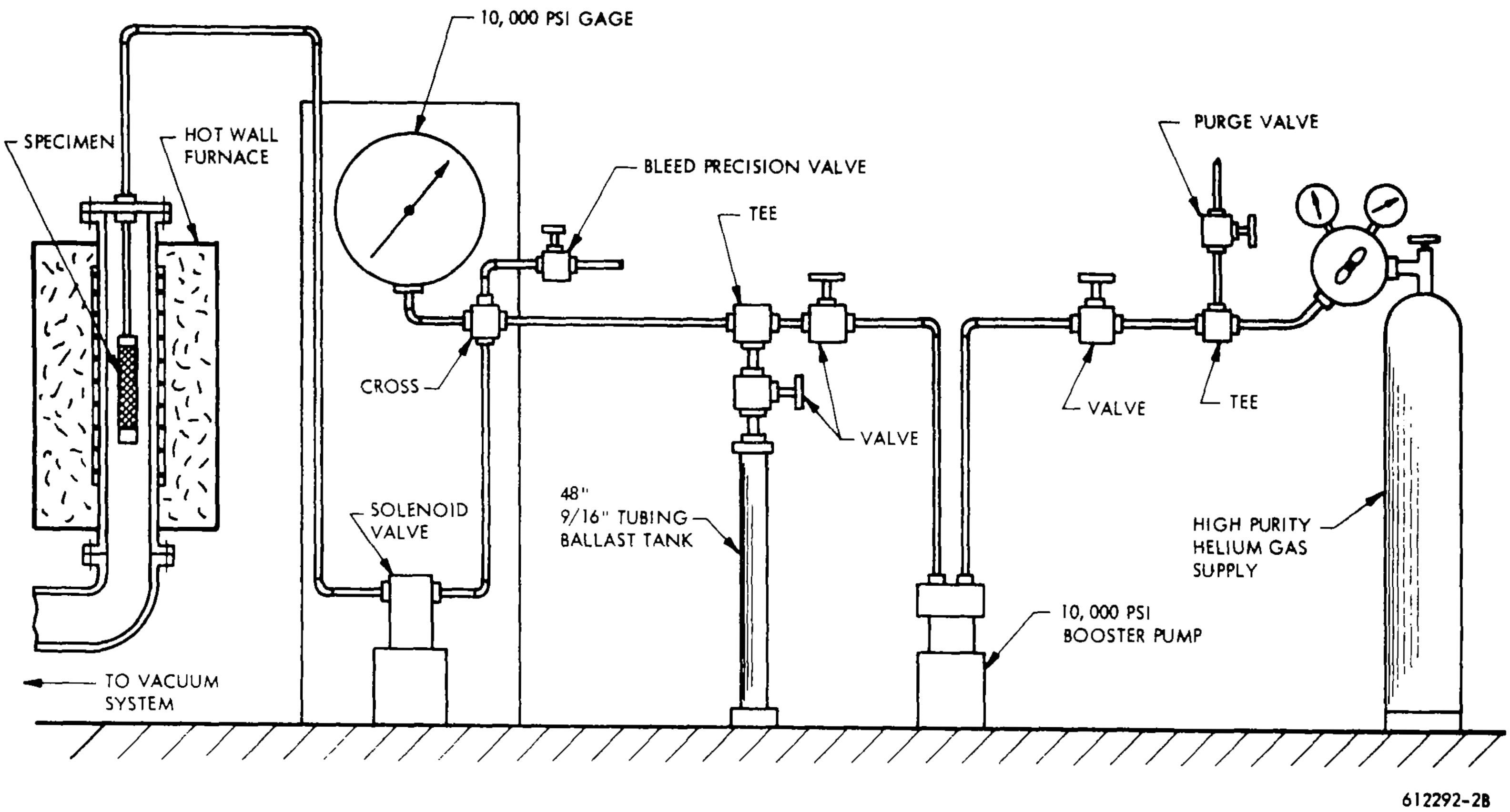


FIGURE 21 - 10,000 psi Pressure Test Rig Schematic

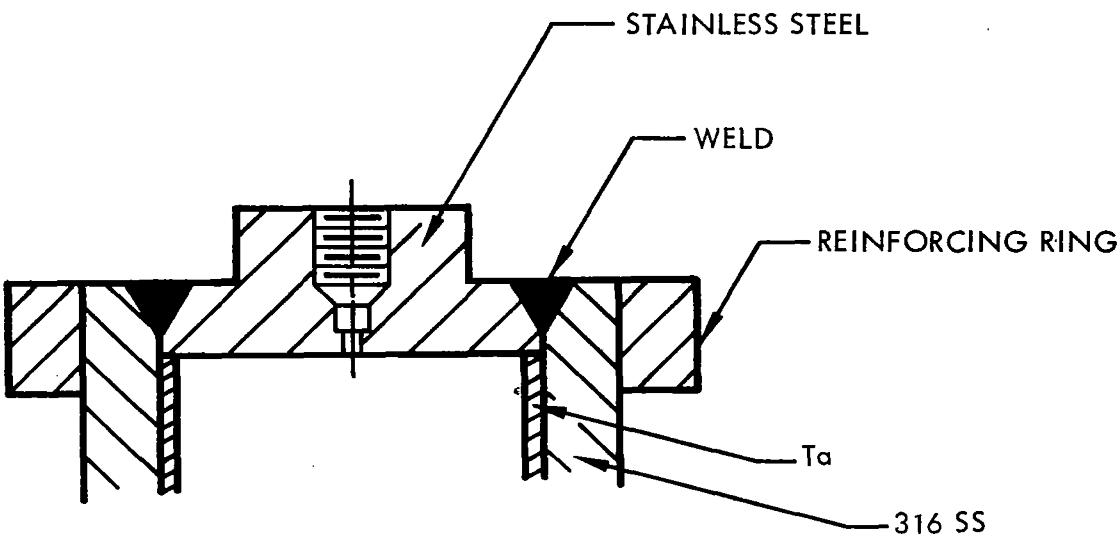


FIGURE 22 - Final End Plug Design

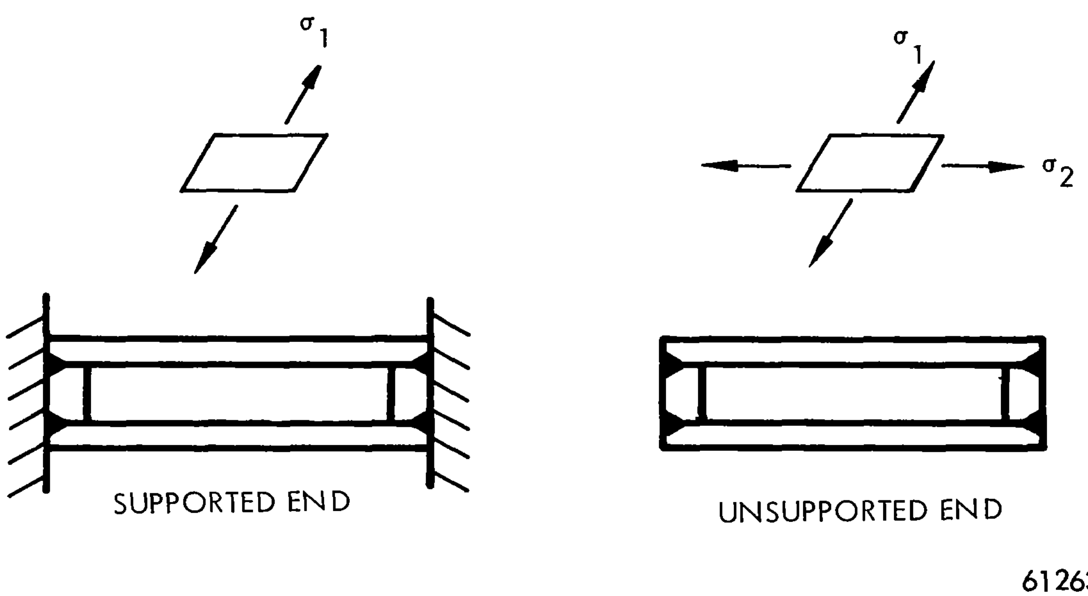


FIGURE 23 - Stress States in Supported and Unsupported End Cylinder Pressure Tests

P = internal pressure in psi, r = mean tube radius, and t = wall thickness. Two types of pressure tests were evaluated; burst tests and creep burst tests, both at 1350°F (732°C). Considerable difficulty was experienced with interrupted tests because of end cap failures and although an improvement was obtained with the design shown in Figure 22, the modification still produced difficulties with the longer time creep burst tests. A tabulation of the burst test data is in Table 10.

<u>Burst Tests.</u> Two burst tests were run to completion and the results are compared with the tensile properties of 316 SS and tantalum.

Tube	Effective Burst Stress (psi)	(732°C) 1350°F 316 SS UTS (psi)	(732°C) 1350°F Tantalum UTS (psi)
A-3 (Mandrel Extruded)	38,700	30,000/50,000	27,000 - Annealed[9] 40,000 - Stress Relieved[10]
B-1 (Filled Billet Extrusion)	30,100	30,000/50,000	As Above

Although the comparison of a single test may be misleading the apparent higher strength of the Lot A (Mandrel Extruded) tubing over Lot B (Filled Billet Extrusion) is most likely caused by the unannealed tantalum in Lot **A.** The higher hardness and stress relieved structure of the tantalum liner of Lot A was previously noted. Since both the tantalum liner and the stainless steel contribute to the overall strength of the bimetal tubing, the strength levels observed fall within the range expected for a 1350°F (732°C) composite. The heavier wall section of the Lot C tubing, (0.104 inches compared to approximately 0.085 inches) reduced the effective wall stress and prevented a burst test comparison on the 10,000 psi capacity apparatus.

TABLE 10

TUBE BURST AND CREEP BURST TEST RESULTS

Tube No.	Date	Specimen Description	Internal Pressure psi	Effective Stress σ_e psi (2)	Time Hours To Failure	Total Strain %	Failure Mode
C-1	7-21-67	As fab. End plug No. 1	6340	22,700	Burst Test	0.00	Gas Fitting
B-1	10-20-67	As fab. End plug No. 2	7000	30,100	Burst Test	6.1	Rupture
A-1	2-23-68	Tandem Test, As Fab., End Plug No. 1	7350	30,200	Burst Test	1.0	End Cap
C-2	2-23-68		7350	26,500	Burst Test	0.00	Did not Fail
C-2	2-26-68	Re-run at Higher Pressure	8900	32,100	Burst Test	0.23	End Cap
A-2	3-6-68	As fab. End plug No. 1	8000	32,900	Burst Test	0.49	End Cap
A-3	3-19-68	As fab. End plug No. 2	9100	37,400	Burst Test	1.96	Inlet Tubing
A-3	3-20-68	As above, re-run	9420	38,700	Burst Test	5.50	Rupture
C-3	7-30-68	As fab. End plug No. 2	9800	35,400	Burst Test	0.11	End Cap
A-6	4-10-68	Tandem Test End plug No. 2 - Heat Treated (1)	4250	17,500	18.9	9.1	Rupture
A-5	4-10-68	- As Fab.	4250	17,500	18.9	0.00	Did not Fail
A-5	4-16-68	As above, re-run	4250	17,500	415	2.59	End Cap
C-6	6-3-68	Tandem Test End plug No. 2 - Heat Treated (1)	5600	20,200	7.6	6.1	Rupture
C-4	6-3-68	- As Fab.	5600	20,200	7.6	0.23	Did not Fail
C-4	6-10-68	As above, re-run	5600	20,200	21.0	5.20	Rupture
C-5	6-11-68	As fab. End plug No. 2	4250	15,300	158	0.69	End Cap

$\sqrt{3}/2$ r/t

Values:

Type A = 4.11

Type B = 4.31

Type C = 3.61

(1) Heat Treated at 1650°F for 300 Hours to produce a 0.2 mil diffusion zone. Bimetal bond intact prior to pressure test.

(2) $\sigma_s = \sqrt{3}/2 \; r/t \; P$

r = mean radius
t = thickness
P = internal pressure psi

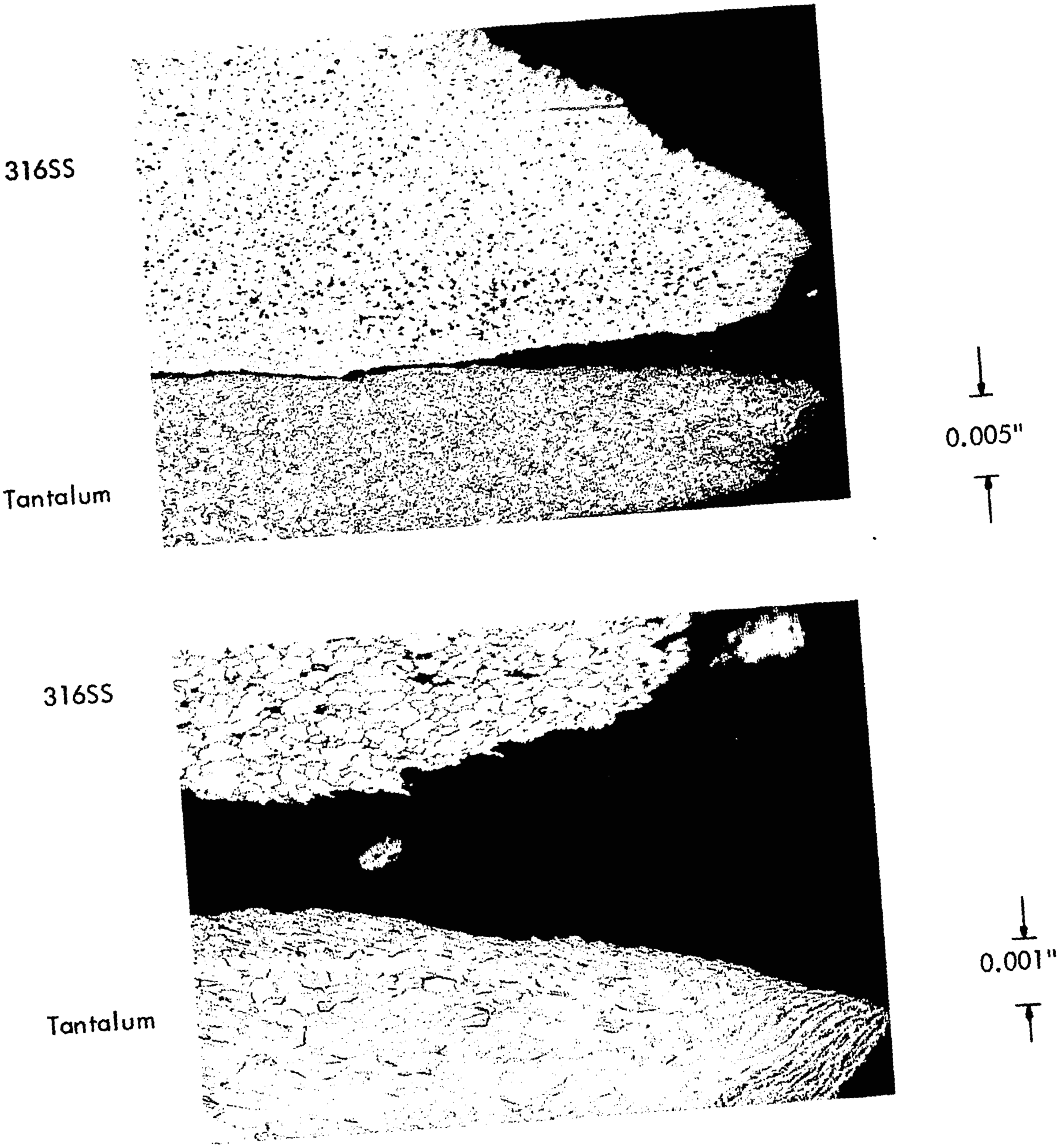
The Lot A and Lot B specimens exhibited over 5% diametral strain before failure. As shown in Figure 24, a fracture section of the Lot A tubing, the reduction in area of the tantalum component exceeded 90%. The 316 SS generally exhibited less than 50% reduction in area and intergranular failure.

Creep Burst Tests. The results of the creep burst tests are listed in Table 10 and plotted in Figure 25. Typical stress rupture properties are included on Figure 25 for 316 SS and tantalum which indicate that the tubing specimens exhibited strength properties within the expected range.

Two of the rupture specimens, A-6 and C-6 were heat treated for 300 hours at 1650°F (899°C) to produce a 0.2 mil interdiffusion zone. The heat treatment markedly lowered the strength of both specimens. The tests on specimens C-5 and A-5 were prematurely interrupted because of end cap failures. Sufficient data is not available to make a direct comparison of the creep burst strength properties of the three types of tubing. The data indicate, however, that the bimetal tubing strength properties may be reliably predicted from the base metal component properties.

The fracture areas of the creep burst tested specimens are similar to the short time fractures as shown in Figure 26. The tantalum component failed by shear with over 90% reduction in area. The duplex microstructure of the 316 SS component evident in Figure 26 will be discussed later.

In summary, the tubing burst tests indicate that the as-fabricated tube strength properties may be predicted from component properties. The 1650°F (899°C) treatment reduced the strength levels of Lot A and Lot C tubing.



Specimen A-3, Burst Stress 38,700 psi tested as fabricated at 1350°F (732°C)

FIGURE 24 - Fracture Edges of Burst Tested Lot A, Mandrel Extruded, Bimetal Tubing

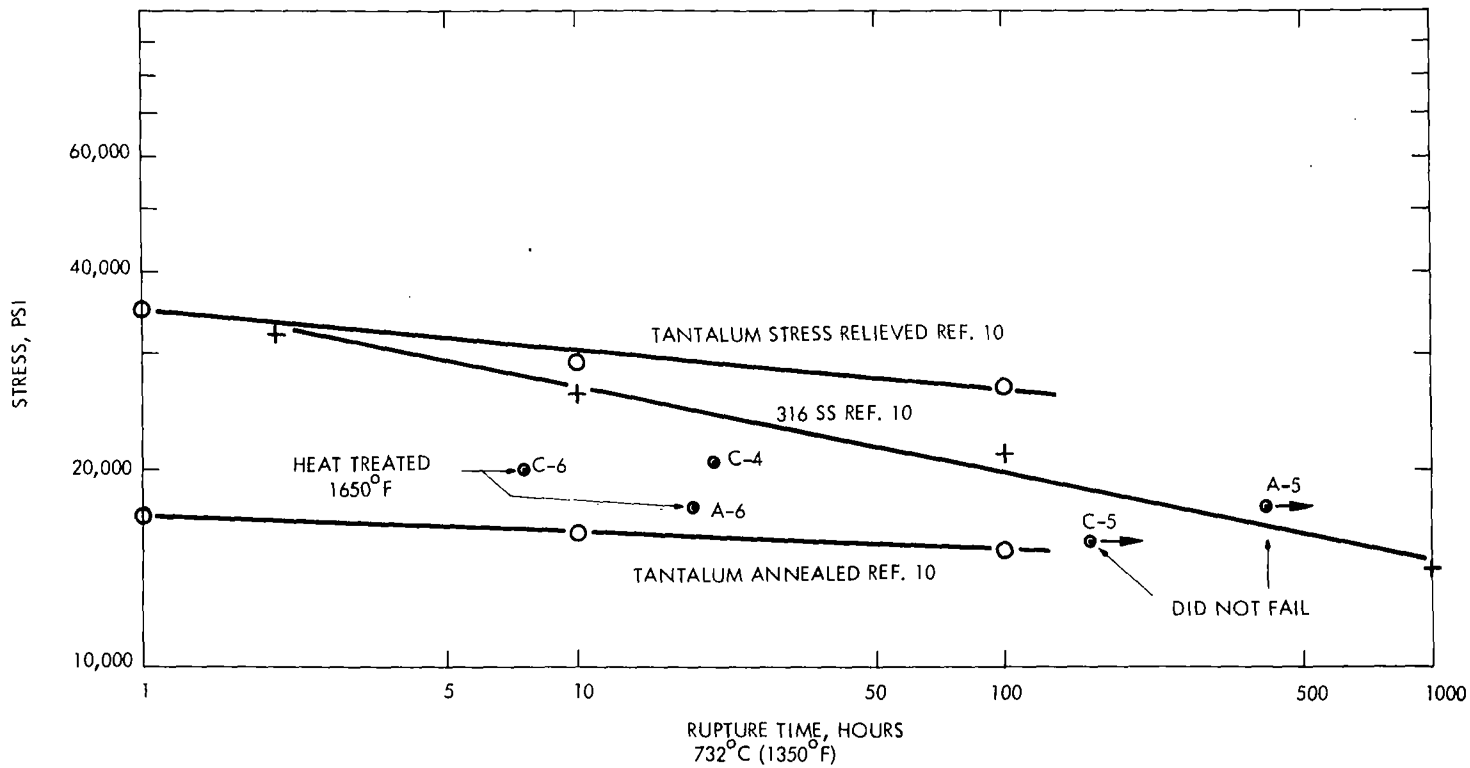
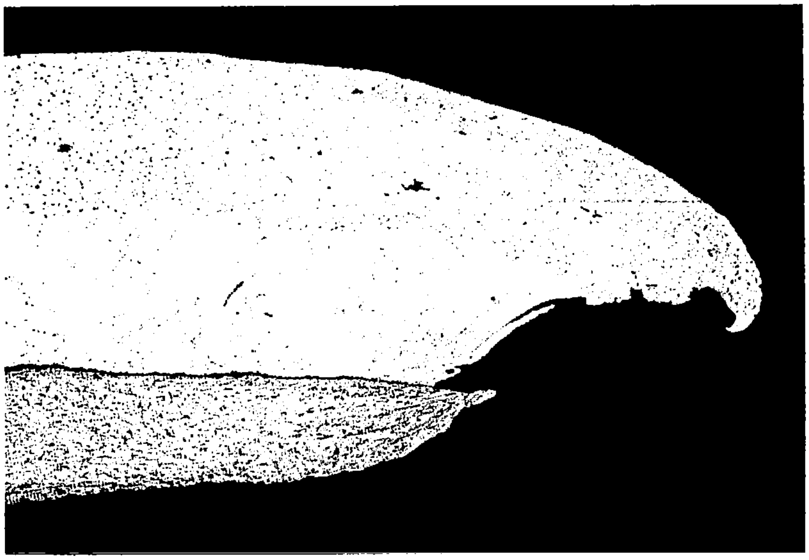


FIGURE 25 - Stress Rupture Properties of Bimetal Tubing

316SS

Tantalum



0.010"

50X

(a) Overall Failure Area Showing Transgranular Ta Failure and Intergranular 316 SS Failure

0.001"

400X

(b) Intergranular 316 SS Failure

Specimen A-6, 17,500 psi stress - 9.1 hours 1350°F (732°C)

FIGURE 26 - Fracture Areas of Creep Burst Tested, Type A Mandrel Extruded Bimetal Tubing

G. THERMAL CYCLE TESTING

Thermal cycle tests were performed on specimens from all three lots of tubing. The test provides a qualitative measure of the ability of tubing specimens to withstand the thermal excursions expected in service. The apparatus used for thermal cycle testing is shown schematically in Figure 27 and a detailed description is given in Reference 2. Originally, the thermal cycle was to consist of heating the specimen to 1350°F (732°C), holding for 5 minutes and cooling to 600°F (316°C) within 30 seconds. This cycle was to be repeated a total of 20 times. After six specimens had been tested in the above manner, the test requirements were changed so that the holding time at 1350°F (732°C) was increased to 60 minutes for each cycle and the total number of cycles was increased to 100. In order to accommodate these changes and to complete the testing as quickly as possible the apparatus was adapted for automatic operation. The fully automated system shown in Figure 28 provides a considerable improvement over the manually operated tests both from the standpoint of consistency and economy.

The major stress contribution during the thermal cycling test is from the marked difference in thermal expansion characteristics of the 316 SS and tantalum, approximately 10.3×10^{-6} inches/inch/°F for 316 SS and 3.6×10^{-6} inches/inch/°F for tantalum. In addition the intermetallic layer produced in the 1650°F (899°C) heat treated specimens further confounds the stress situation. The major anticipated effect of the thermal cycle exposure would be a rupture of the bimetallic bond. Ultrasonic testing and destructive testing was used to measure the post thermal cycle bond integrity.

A 2 inch specimen length was generally used which was sufficiently long to minimize end effects of the bimetal stress field. Since the Lot C, explosively bonded, tubing contained spacing protrusions, each with an associated unbond area, two different types of spacing configurations were evaluated. The first consisted of three equally spaced protrusions in the center of the specimen and a second type evaluated a helical spiral spacing with one

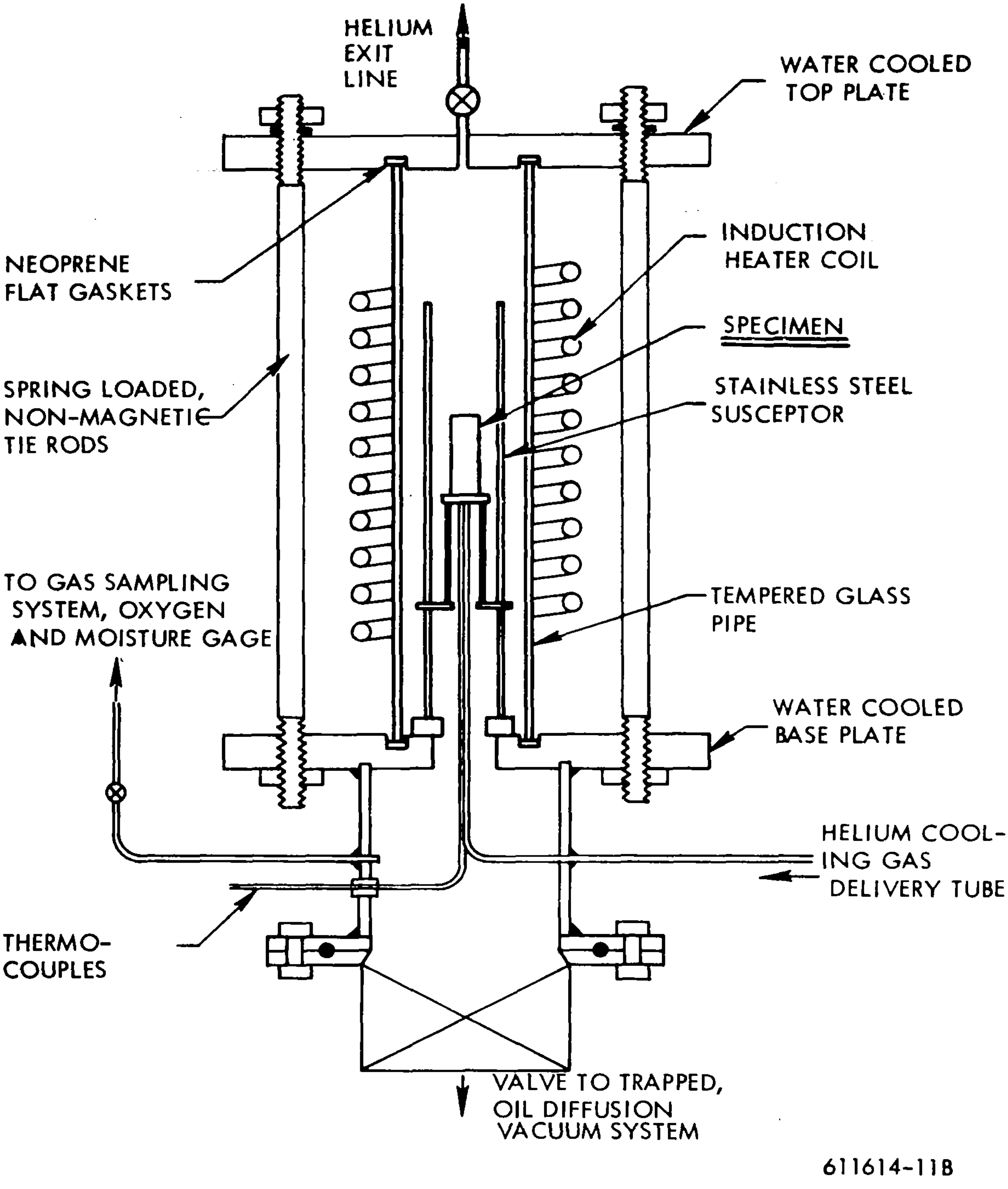


FIGURE 27 - Schematic of Thermal Cycle Test Apparatus

FIGURE 28 - Automated Thermal Cycle Apparatus

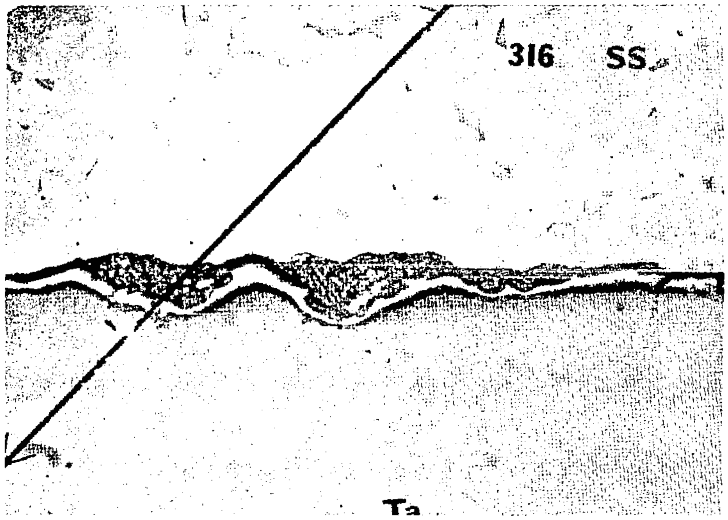
protrusion at either end of the specimen. Explosively bonded tubing sections without a spacing protrusion were also evaluated.

Thermal cycle tests were performed on as-fabricated specimens and on specimens which were heat treated at 1650°F (899°C). The heat treating was performed under ultra-high vacuum conditions of 1 x 10^{-7} torr in a bakeable, metal-sealed system. The samples were wrapped in Ta foil and contained within a Vycor tube which was heated by a hot wall furnace. A Pt/Pt-13Rh thermocouple embedded in the load was used to monitor the specimen temperature which was maintained at ±5°F. The specimens were ultrasonically, dye-penetrant, and metallographically inspected before and after annealing to determine if any changes occurred during annealing. The thermal exposure of the test specimens produced diffusion zones as predicted except in the Lot C, explosively bonded tubing. The growth was irregular in Lot C as shown in Figure 29a. Both Lot A and Lot B tubing produced diffusion zones as shown in Figure 29b.

During the testing of specimens B-3, B-5, and A-4, the gas outlet valve failed to seal properly and allowed an air leak to occur. This caused oxidation of both Ta and 316 SS, as well as severe specimen warpage and invalidated the test.

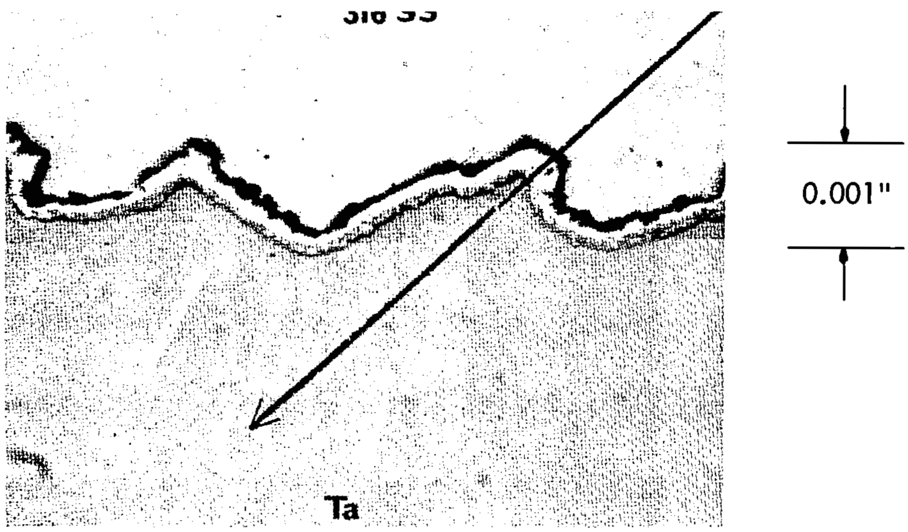
The results of the thermal cycle testing are tabulated in Table 11. Both types of extruded tubing, Type A and B, survived the thermal cycle tests and 1650°F (899°C) pre-treatment with only one case of bond deterioration. The single unbond area, in specimen B-4, was present in the as-fabricated specimen and may not be representative of the tubing in general. The more severe thermal cycle sequence including 100 cycles and a 60 minute hold at 1350°F (732°C), was used for all of the Type A specimens.

The explosively bonded tubing, Lot C, showed a significant growth in the spacer unbond areas after the thermal cycle test both in the as-received and heat treated conditions. Neither type of spacer design, the three spacer single plane or the helical spiral was effective in preventing additional unbond propogation during thermal cycling. The section of



540X

(a) Lot C Explosively Bonded Tubing, Heat Treated at 1650°
for 300 Hours.



540X

(b) Lot B_6 Filled Billet Extrusion, Tubing Heat Treated at
1650°F for 300 Hours

FIGURE 29 - Interdiffusion Zones in Ta/316SS, Bimetal Tubing

TABLE 11

RESULTS OF THERMAL CYCLE TESTING ON BIMETAL TUBING

Specimen No.[2]	Condition[1]	Length	No. Cycles	1350°F Hold Time - Minutes	Pre Test Comments	Results
A-1	As fab.	2"	100	60		No unbond
A-2	As fab.	2"	100	60		No unbond
A-3	Heat Treat.	2"	100	60		No unbond
A-5	Heat Treat.	2"	100	60	*Specimen dented*	No unbond
B-1	As fab.	5"	40	5		No unbond
B-2	As fab.	1-3/4"	100	5		No unbond
B-4	As fab.	2"	100	60	Unbond area as fabricated.	Unbond - Pre test area grew significantly.
C-1	Heat Treat.	2-3/4"	20	5	3 spacers in same plane - unbond at spacers grew during heat treat.	No change in size of pre test spacer unbond.
C-2	Heat Treat.	2-3/4"	100	5	3 spacers in same plane - unbond at spacers grew during heat treat. Additional small unbond area.	Unbond - Pre test area at spacers and additional area grew.
C-3	Heat Treat.	2-3/4"	100	5	3 spacers in same plane. Unbond at spacers grew during heat treat. Continuous bimetal intermetallic.	Unbond - Spacer unbond areas connected to form large unbond area. Intermetallic layer fractured.
C-4	Heat Treat.	2-3/4"	100	5		
C-5	As fab.	2"	100	60	2 helical spacers. Unbond at spacers.	Unbond - Spacer unbond areas grew.
C-6	As fab.	1/2"	100	60	No spacers	No unbond
C-7	As fab.	2"	100	60	2 helical spacers, Unbond at spacers. Continuous bimetal intermetallic.	Unbond - Spacers grew slightly - Intermetallic layer fractured.
C-8	Heat Treat.	2"	100	60	2 helical spacers, Unbond at spacers grew slightly during heat treatment.	Unbond - Spacer unbond grew to large area.

(1) Condition - As fabricated or heat treated at 1650°F (899°C) for 300 Hrs.

(2) Type A Mandrel Extruded
Type B Filled Billet Extrusion
Type C Explosively Bonded

explosively bonded tubing which was free of all spacing protrusions did not unbond during thermal cycling. The intermetallic layers present in the as-fabricated and heat treated specimens were fractured extensively in both longitudinal and transverse directions during thermal cycling.

The test results indicate that either type of extruded tubing has good bimetal bond durability during thermal exposure and thermal cycle testing. Explosively bonded tubing, conversely, suffers significant unbonding unless the "built-in" defects at the spacer protrusions are eliminated.

H. METALLOGRAPHIC, CHEMICAL, AND MICROPROBE ANALYSES OF HEAT TREATED AND THERMALLY CYCLED SPECIMENS

Interstitial Chemical Analysis. Chemical analyses for C, O and N were performed on heat treated, and thermally cycled Lot A specimens which contained a 0.2 mil diffusion zone. The analytical results are summarized and compared to the as-fabricated analyses in Table 12.

Heat Treatment Effects. The extended service simulating heat treatment of 1650°F (899°C) for 300 hours produced a marked change in interstitial content of both the tantalum and 316 SS components. In general, the carbon, oxygen and nitrogen content of stainless steel decreased and the oxygen and nitrogen content of the tantalum increased. Since the heat treatment was accomplished in an ion pumped system at less than 1×10^{-7} torr, the interstitial composition changes are assumed to be produced by diffusion across the bimetal interface. The free energies of the possible interstitial-metal compounds in 316 SS at 1650°F (899°C) indicate that TaC, TaN and Ta_2O_5 would all be formed at the expense of carbon, nitrogen and oxygen in solid solution or as a precipitate in the iron base 316 SS. Since the interdiffusion zone between the tantalum and 316 SS was removed in preparation for chemical analysis, a complete material balance could not be made. The formation of tantalum carbide and/or nitride compounds in the interdiffusion zone could not be confirmed by the electron

TABLE 12

INTERSTITIAL CHEMICAL ANALYSES OF TYPE A (MANDREL EXTRUDED) TUBING

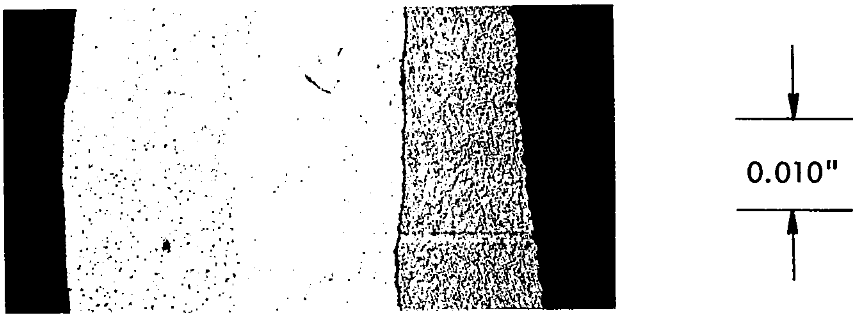
Analyses	As Fabricated	1650°F (899°C) - 300 Hr. at <1 x 10^{-7} Torr Heat Treatment		
	Analyses in Parts per million by weight			
Tantalum				
C	39		30	
O	53		110	
N	28		120	
316 SS		Course Grained Area	Entire Specimen	Fire Grained Area
C	700	140	130	130
O	70		29	
N	380		250	260

beam microprobe since this analytical technique was insensitive to nitrogen or carbon. The greatest change in tantalum interstitial level was for nitrogen which increased from about 30 ppm to 120 ppm. Oxygen content in the tantalum doubled from 50 ppm to 110 ppm. No significant change was observed in the tantalum carbon content throughout the testing program including thermal cycling.

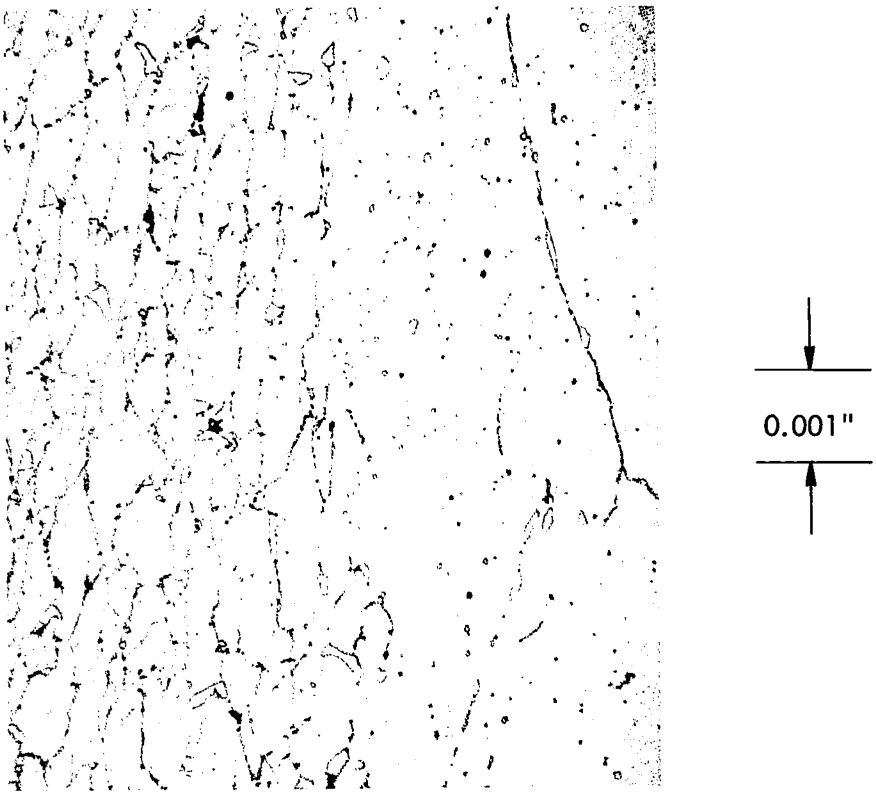
The 316 SS carbon content decreased from 700 ppm to approximately 130 ppm. Since an increase in tantalum carbon content was not observed, it is assumed the carbon diffused to form a metal-carbide interdiffusion layer. Both the oxygen and nitrogen content also decreased following heat treatment which may be attributed to diffusion to the refractory metal.

Thermal Cycling Effects. Following the thermal cycle tests which involved over 100 cumulative hours of 1350°F (732°C) exposure to 1×10^{-5} torr vacuum or high purity helium (which at 1 ppm total impurity level is roughly equivalent to a 1×10^{-4} to 1×10^{-3} torr vacuum) the tantalum bimetal component showed an additional increase in oxygen content from 110 ppm to 300 ppm which is assumed to come from the test environment. The 316 SS showed an increase in carbon and oxygen content (70-100 ppm). Again this increase may be caused by contamination from the thermal cycling atmosphere.

Metallographic Correlation. An examination of sections from the bimetal tubing following heat treatment for 300 hours at 1650°C, Figure 30, Lot C tubing and Figure 26, Lot A, indicates an increase in the 316 SS grain size adjacent to the tantalum. If carbide and nitride precipitates are assumed to provide a degree of grain size stabilization in the 316 SS at 1650°F, the large grained area adjacent to the tantalum may be assumed to be depleted in carbon and nitrogen precipitates. Additional examinations of the coarse grained area dimensions as a function of time indicate the area increased with the square root of time which corresponds to a diffusion phenomena.



(a) Dual Grain Size 50X



(b) Dual Grain Size and Grain Boundary Precipitate 500X

FIGURE 30 - Large Grain Size 316SS in Heat Treated Lot C, Explosively Bonded, Bimetal Tubing

Assuming that the large grained area in 316 SS was due solely to nitrogen diffusion depletion, an exploratory calculation was made on the large grain area dimensions as a function of time. Smith[12] has derived an equation describing the behavior postulated and it is shown schematically in Figure 28. The equation, solved for the diffusion coefficient is

$$\frac{(C_i - 2/3C_B)}{2} \frac{\partial x^2}{\partial t} = DC_B$$

where C_i is the concentration of N in the material, C_B is the solubility limit of N in the material, and $\frac{\partial x^2}{\partial t}$ is the parabolic rate constant squared.

Substitution of the parabolic rate constant for growth of the coarse-grained zone, the N content of the heat treated 316 SS, and the solubility limit of N in stainless steels at 1600°F (899°C) gives a value for D of 3×10^{-8} cm^2/sec. The literature gives values of 3 to 8×10^{-8} cm^2/sec for the diffusion coefficient of N in stainless steel.

Hardness Changes. A comparison of the before and after heat treating hardness traverses of the tantalum and stainless steel is shown in Figures 31 and 32. In general the tantalum increased in hardness following the thermal exposure with an increasing hardness gradient towards the bimetal interface. The hardness gradient may be assumed to follow an interstitial content gradient although this was not confirmed by chemical analysis. The Lot A tubing showed a drop in tantalum hardness, but this is due to stress relieving of the worked tantalum. The 316 SS decreased in hardness following heat treatment especially the explosively bonded tubing in which the stainless steel was severely hardened by the explosive deformation process.

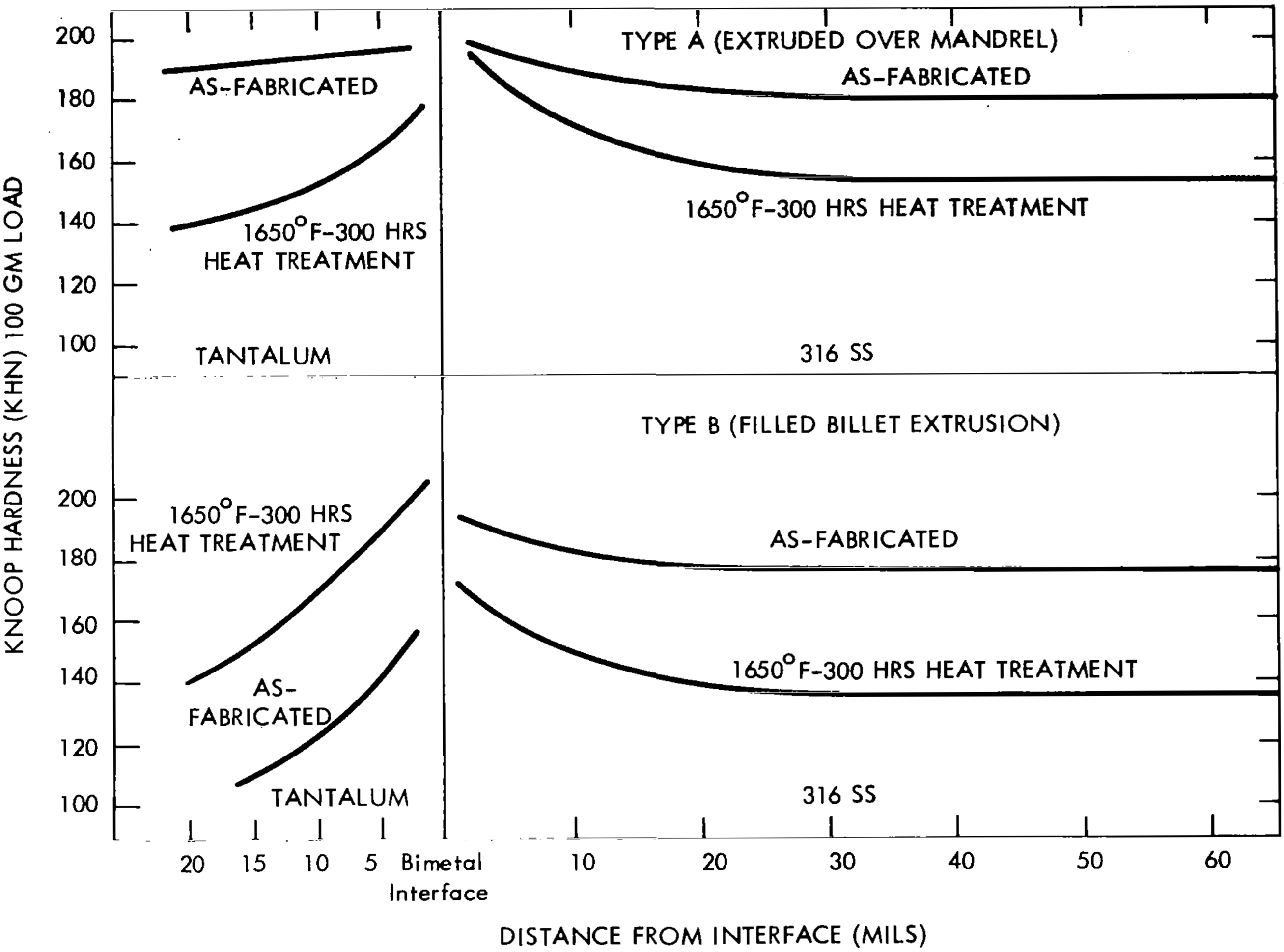


FIGURE 31 - Hardness Traverse Comparison of Heat Treated Lots A and B, Bimetal Tubing

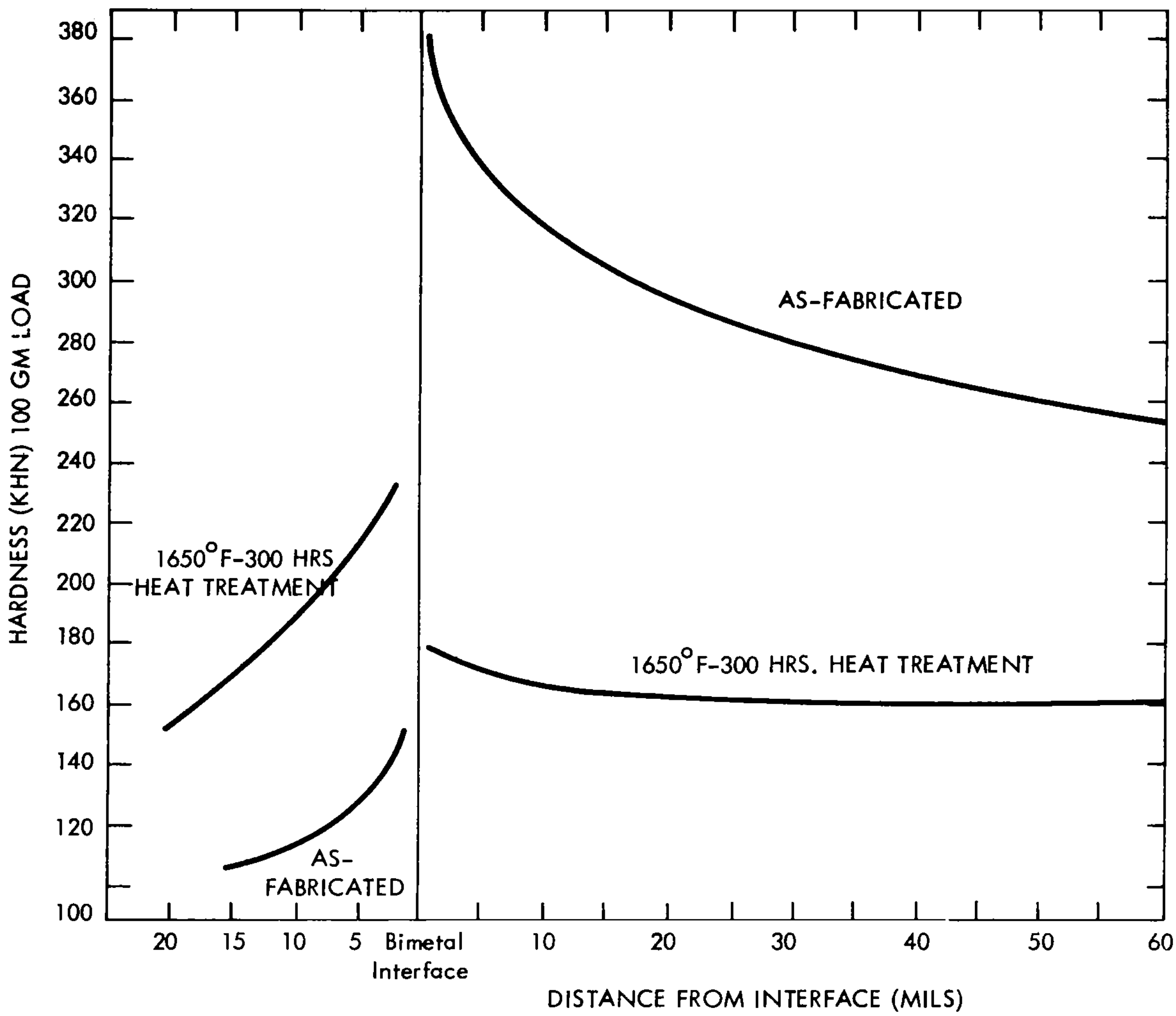


FIGURE 32 - Hardness Traverse of Heat Treated Type C Tubing

Microprobe Analyses. Heat treated, 1650°F (899°C) - 300 hours, specimens of Lot B (filled billet extrusion) and Lot C (explosively bonded) bimetal tubing were analyzed by the electron beam microprobe. Figure 29 shows the two samples analyzed and the direction of the interface traverse is indicated by the arrow. The light layers at the interdiffusion zone were found by both point and line traverse analyses, to be 92% tantalum in the Lot C (explosively bonded tubing) and 90% tantalum in the Lot B (filled billet extruded) tubing. The large irregular dark zones in the Lot C tubing contained Ta and Ni, Fe, and Cr in approximately the same ratio as in stainless steel.

The line and point traverses showed no evidence of interdiffusion of tantalum and the stainless steel components beyond the metallographically observable intermetallic layer.

VI. CONCLUSIONS

1. Bimetal tubing of acceptable dimensional properties can be made by explosive bonding and two techniques of high temperature coextrusion.

2. The explosively bonded tubing contains inherent unbond areas at the spacing protrusions required for the bonding process.

3. The spacer unbond areas in the explosively bonded tubing propogate under subsequent thermal exposure and/or thermal cycling.

4. The thermal expansion characteristics of the three lots of tubing are essentially the same and are controlled by the thicker layer of high expansion rate 316 stainless steel. The coefficient of expansion of 316 stainless steel can be used to predict the diametral and longitudinal thermal expansion characteristics of the bimetal tubing. However, changing the ratio of stainless steel to tantalum would probably change the thermal expansion characteristics.

5. Pressure burst tests of the bimetal tubing at 1350°F (732°C) did provide an accurate comparison of the tubing performance. The strength levels of two tubes that were tested to failure indicated an ultimate tensile strength of 30,000 to 38,000 psi. This compares to tensile strength values of 30,000 to 50,000 psi for 316 SS at 1350°F.

6. Type A, mandrel extruded, and Type C, explosively bonded tubing was creep burst tested at 1350°F (732°C). Tests to failure were obtained at relatively high stress levels, 17,000 psi to 20,000 psi, and short times of less than 20 hours. The creep rupture data agree well with data for 316 SS. Pre-test heat treated specimens (1650°F-300 hours) were significantly weaker than the as-fabricated specimens most likely because of the stress relief of the stainless steel and tantalum components during the heat treatment. As in the short time burst tests, the strength performance of the bimetal tubing could be predicted from 316 SS data.

7. Thermal cycle tests of the as-fabricated and heat treated tubing indicated the bond durability of the two types of extruded tubing was superior to the explosively bonded tubing. The explosively bonded tubing suffered severe propogation of inherent spacer unbond areas during both the thermal exposure and during the thermal cycle test. A section of explosively bonded tubing purposely prepared to contain no unbonded areas survived the thermal cycle test intact.

8. Chemical analysis of a specimen heat treated to simulate time at temperature service conditions indicated a significant migration of nitrogen and oxygen from the 316 SS component to the refractory metal. Hardness measurements of the contaminated tantalum, however, indicate that acceptable room temperature strength and ductility were maintained. Also, all of the failed burst test specimens showed extremely ductile failures in the tantalum component.

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