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MERCURY AMALGAM BONDING OF URANIUM AND COPPER

L. G. GLASGOW

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
CARBIDE AND CARBON CHEMICALS DIVISION
UNION CARBIDE AND CARBON CORPORATION
POST OFFICE BOX P
OAK RIDGE, TENNESSEE

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Written by: L. G. Glasgow

Work done by: L. G. Glasgow
J. N. Hix
C. D. Smith

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OAK RIDGE NATIONAL LABORATORY
Operated by
CARBIDE AND CARBON CHEMICALS DIVISION
Union Carbide and Carbon Corporation
Post Office Box P
Oak Ridge, Tennessee

[redacted]

[redacted]

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ABSTRACT

Data are presented relating strength of silver-mercury bonds between uranium and copper, copper and copper, and copper and silver to bonding time and temperature. Bonding time was varied from 1/2 hour to 6 hours; temperature was varied from 350°C to 550°C. In general strength increased with increasing bonding time and with increasing temperature. Machined uranium samples were compared to as-rolled uranium samples, machined samples gave superior strength values.

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INTRODUCTION

This report is a summary of one phase of work done to develop methods of bonding uranium to a metal which is a good conductor of heat. The requirements of the bond are that it have a fair tensile strength and be a fair heat conductor. Saller and Keeler[1,2] developed a method for bonding uranium inside nickel tubes by plating the surfaces to be joined with silver and diffusion bonding the mating surfaces with the aid of an amalgamating agent such as mercury. Silver was chosen for contact with uranium because it is easy to plate on uranium and because there is no danger of brittle Ag-U compounds being formed at elevated temperatures.[1,3]

SUMMARY

Preliminary plating experiments were carried out to determine the best conditions for silver plating and copper plating of uranium. Silver plates were tested for oxidation resistance at 300°C or 350°C as plated and as plated plus cold rolled. The effect of preheating in vacuum before testing for oxidation resistance was determined. Specimens of uranium were prepared by silver plating or copper plating on top of copper replacement coatings obtained by immersion of the samples in $CuSO_4$ solution. These were tested for oxidation resistance at 350°C.

Samples of silver plated copper bonded to silver were prepared by pressing amalgamated surfaces together at temperatures from 350°C to 550°C for time periods of 2 to 6 hours. Bond strength determinations were made. Similar tests were made on silver plated uranium bonded to silver plated copper by pressing amalgamated surfaces together at 350°C for 4 hours. The factor investigated in this series of tests was the effect on the plating bond of vacuum annealing silver plated uranium prior to amalgamation.

Bond strength tests were made on samples of silver plated copper bonded to silver plated copper by pressing amalgamated surfaces together with a hydraulic press equipped with electrically heated platens at 350°C and 6,100 psi pressure for time periods of ½ hour to 2 hours.

CONCLUSIONS

On the basis of the data obtained in the present work, the following conclusions may be made:

(1) Silver plating on uranium does not afford good oxidation protection at temperatures of 300 to 350°C.

[redacted]

(2) Bond strength of mercury amalgamated silver increases with bonding temperature at constant time and with increasing time at constant temperature using times up to 6 hours and temperatures up to 550°C.

(3) The effect of vacuum annealing on the bond strength of a silver plate on uranium is not known since this bond is stronger than the silver-amalgam-silver bond.

(4) Machined surfaces on uranium produce considerably stronger bonds than as-rolled surfaces when the surface is silver plated and joined to silver plated copper by mercury amalgamation.

(5) Bond strengths as high as 4455 psi can be obtained in sandwiches of silver plated uranium joined to silver plated copper with a mercury amalgam bond.

(6) Bonds as strong as 6000 psi were obtained by amalgamating silver plated copper plates and pressing together at 350°C and 6100 psi.

EXPERIMENTAL PROCEDURE

A. Plating of Uranium.

The principal problem in plating is obtaining a plate which will stand up at elevated temperatures. Silver plate, due to its porosity and capacity for oxygen, affords little protection for uranium when heated in air. The following is a description of the methods used to try to overcome this problem: A surface free of flaws and impurities is essential in obtaining a sound plate; the plate is no better than the surface plated. Difficulty was encountered in plating surfaces of as-rolled specimens even though the surfaces were deeply etched prior to plating. Indications are that plating on rolled surfaces does not adhere as well as to machined surfaces. The procedure used to silver plate uranium is listed in the appendix. 2 x 3/4 x 1/8 inch uranium samples sheared from 1/8 inch plate were used in the tests. Uranium plated with 0.003 inches of silver and placed in an air atmosphere muffle furnace at 300°C cracked at the edges and blistered on the flat surface to the extent that within two hours, the plating was completely separated from the heavily oxidized uranium. Vacuum annealing at 600°C for 1 hour before heating in air did not improve the results.

Specimens plated to 0.003 inches and given a 0.002 inch reduction in a hand roll were greatly improved. (4) Although rolling did not reduce the porosity noticeably, it improved adhesion so that although the plating cracked at the edges, oxidation did not proceed sufficiently far to cause spalling of the plate. The size of blisters due to porosity was greatly reduced. If silver plated uranium were bonded to another wrought metal, the bonded area would not be exposed to air; therefore, porosity of the plate was considered no problem in the actual bond. Vacuum annealing at 600°C for one hour after rolling did not improve results. Reduction of the plating current from recommended 25 amps/sq.ft. to 10 amps/sq.ft. did not improve the protective qualities of the plate.

Five specimens were plated using the following procedure:

(1) 2 x 3/4 x 3/16 inch uranium blocks were machined from 1/2 inch thick plate rolled from 1 inch ingot, then annealed.

(2) Edges and corners were rounded to 1/64 inch radius.

(3) Specimens were plated with 0.003 inch silver according to procedure in appendix.

(4) Specimens were polished on buffing wheel.

(5) 0.006 inch additional silver was plated on.

(6) Specimen was reduced 0.001 inch in hand roll.

When these specimens were heated in air at 350°C for 4 hours, the plating cracked at all edges and came completely loose from the four smaller sides of four of the samples. One specimen showed no effects. Similar results were obtained with specimens which were plated with 0.003 inch of silver, reduced 0.002 inch by rolling, were replated with 0.006 inch of silver and given a final reduction, by rolling, of 0.001 inch.

B. Copper Plating.

When uranium was plated with 0.003 inches of copper from a copper sulphate bath, the plate did not adhere to the uranium after heating at 300°C for 1 hour.

0.003 inches of silver was plated over a copper replacement coat obtained on uranium by allowing it to stand in 10% $CuSO_4$ for 24 hours. The specimen was given a 0.002 inch reduction pass. When exposed to air at 350°C, the plating was cracked at the edges and completely separated from the uranium in 1 hour.

A uranium specimen was allowed to stand in 10% $CuSO_4$ for 48 hours and then given a 0.003 inch plating of copper. Over the copper was plated 0.001 inch of silver and the specimen was given a 0.002 inch reduction pass. On exposure to air at 350°C for 3½ hours, the plating did not adhere to the copper.

C. Mercury Bonding of Silver Plated Copper to Silver.

The following tests were run to determine the best conditions for bonding by mercury amalgamation.

Procedure:

(1) 3 x 9/16 x 1/8 inch pieces of machined copper sheet were plated with 0.003 inches of silver in the bath used for uranium and abraded with No. 600 emery paper.

(2) 3 x 9/16 x 1/16 inch pieces of silver were abraded with No. 600 emery paper.

(3) Specimens for measuring relative bond strength were made by overlapping 1 inch of mercury amalgamated surface of a plated copper piece and 1 inch of mercury amalgamated surface of a silver piece (Fig. 1). Excess mercury was wiped off the pieces before joining. These were clamped in vises in sets of four and heated in air.

(4) After bonding the specimens were machined to $\frac{1}{2}$ inch width across the bonded area.

(5) Fig. 1a shows the overlapping of 1 inch used in the strength tests. Since one of the 350°C, 2 hour specimens broke outside the bonded area, the bonded area of the remaining specimens was reduced 50% as shown in Fig. 1b.

Results:

The specimens were pulled in tension in the 120 ton Baldwin Southwark tensile machine. The results of bond tests are listed in Table I. They show a general increase in bonding strength with time at constant temperature and with temperature at constant time. Figs. 2, 3, and 4 show metallographically the improvement of the bond with time at constant temperature and Figs. 5, 6, and 7 show the improvement of the bond with temperature at constant time.

Examination of the fractures of the specimens showed the majority to be at the original interface. Where the fractures included tearing of the silver plating from the copper, the strengths were low indicating that the plate adhesion, not the silver to silver bond, was weak on these specimens.

D. Mercury Bonding of Silver Plated Copper to Silver Plated Uranium.

Pieces of uranium were sheared from a 1/8 inch plate, plated with 0.003 inches of silver, and reduced 0.002 inches in a hand roll. These specimens were bonded to copper plated with 0.003 inches of silver by amalgamating 2 x 3/4 inch surfaces were mercury and heating in a vise. Temperatures below 300°C for periods of from 1 to 6 hours produced in a metallographically poor bond.

To evaluate the bond strength of silver plating on machined and as-rolled uranium, and to determine the effect of vacuum annealing on plate adhesion, relative bond strength tests were run as follows:

Procedure:

3 x 1/2 x 3/16 inch uranium specimens were machined from a 1/2 inch plate rolled from a 1 inch casting. They were plated with 0.003 inches of silver and reduced 0.002 in hand rolls.

3 x 9/16 x 1/8 inch rolled copper sheet specimens were plated with 0.003 inches of silver. Bond strength specimens were made as shown in Fig. 8. Joining surfaces were amalgamated with mercury as plated, and the samples were placed in small vises. The vises were tightened as tight as possible and placed in an air atmosphere furnace at 350°C for 4 hours.

Specimens No. 11, 12, and 13 were prepared by the above procedure.

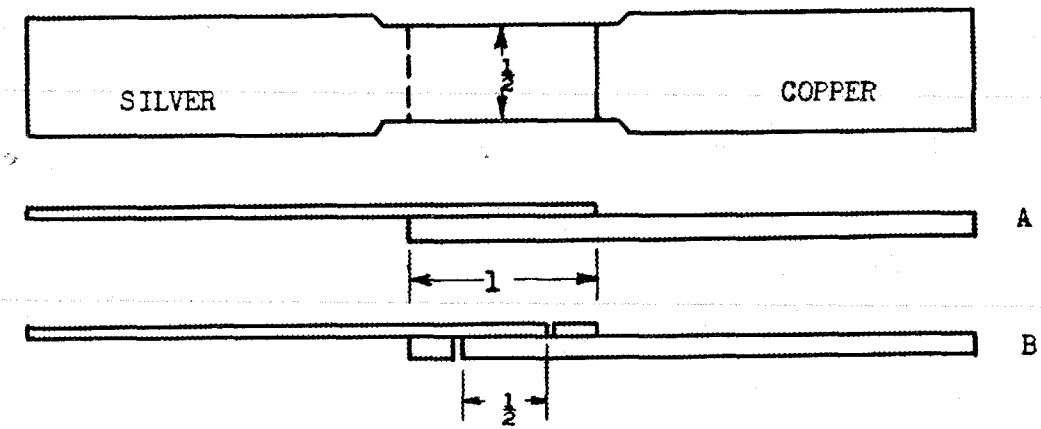


Figure 1

Bond Strength Specimens

Silver Plated Copper to Silver by Mercury Amalgamation Pressure Bonding

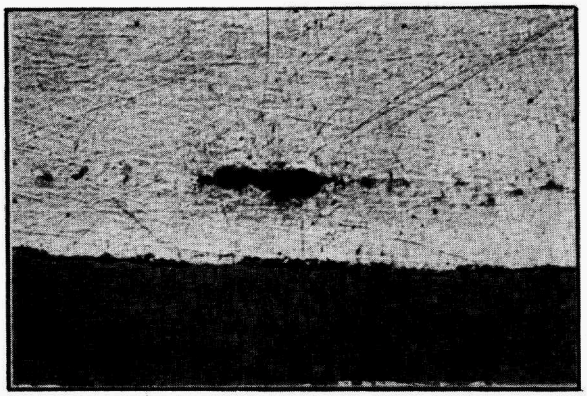
Dwg. No. 10095
Unclassified

TABLE I

Effect of Time and Temperature on Bond Strength of Silver Plated Copper Joined to Silver by Mercury Amalgamation

Temperature °C	Time Hours	Max. Stress Psi	Fracture
350	2	1550	Silver broke outside bonded area.
		1390	Ag to Ag
		1342	Ag to Ag
350	6	1900	Ag to Ag
		1660	Ag to Ag
		2070	Ag to Ag
450	2	1253	Ag to Ag
		970	Ag to Ag
		1210	Ag to Ag
450	4	1538	Ag to Ag
		1210	Ag to Ag
450	6	2100	Ag to Ag
		2140	Ag to Ag
		1245	50% Ag to Ag, 50% Ag to Cu
550	2	1590	10% Ag to Ag, 90% Ag to Cu
		1610	Ag to Ag
		2480	Ag to Ag
550	4	840*	100% Ag to Cu
		1040*	100% Ag to Cu
		1185*	80% Ag to Ag, 20% Ag to Cu
550	6	2420	Ag to Ag
		1850	50% Ag to Ag, 50% Ag to Cu
		2270	Ag to Ag

*Questionable data because of poor plating.



Silver

Original Interface
Silver

Copper

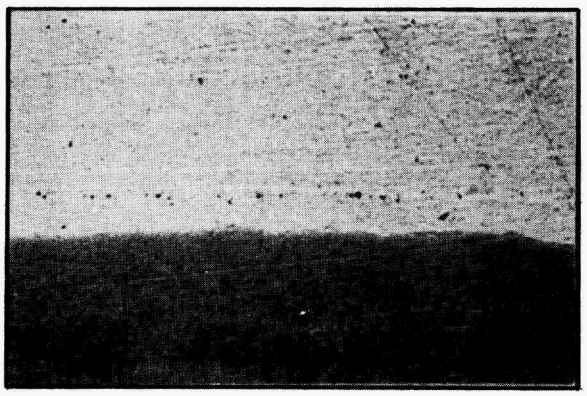
Figure 2 550C 2 hr Y-2483

Silver

Original Interface
Silver

Copper

Figure 3 550C 4 hr Y-2484



Silver

Original Interface
Silver

Copper

Figure 4 550C 6 hr Y-2480

Silver Plated Copper Bonded to Silver by Mercury Amalgamation

Mag.- 400X Unetched

Unclassified

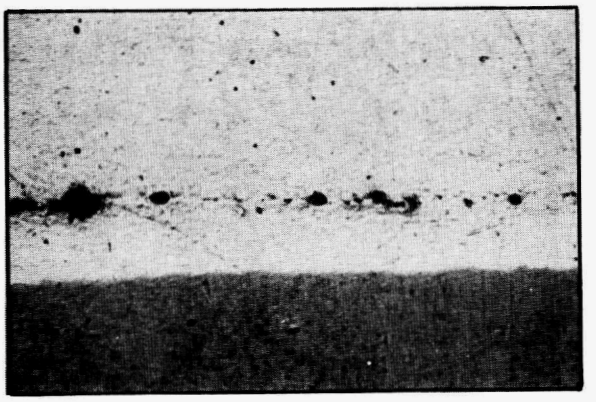


Figure 5 350C 6 hr Y-2482

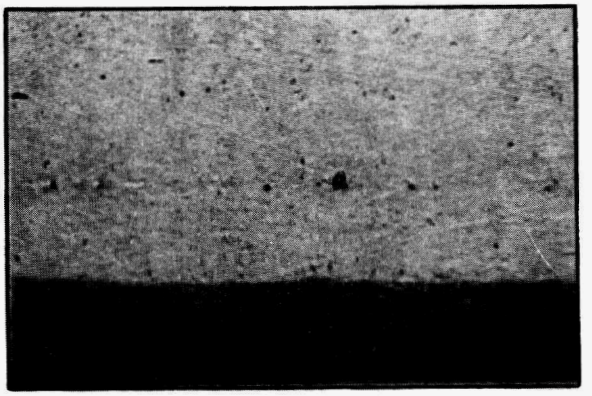


Figure 6 450C 6 hr Y-2481

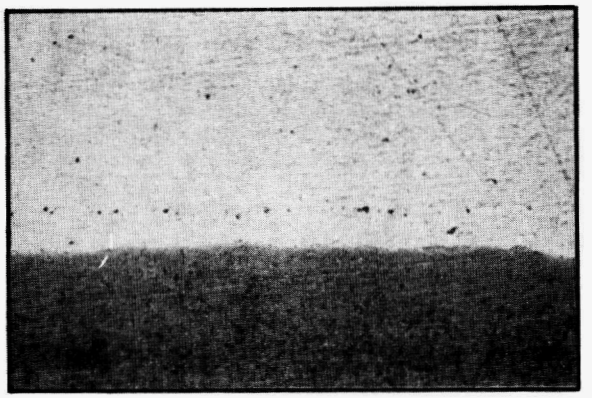


Figure 7 550C 6 hr Y-2480

Silver Plated Copper Bonded to Silver by Mercury Amalgamation

Mag.- 400X Unetched

Specimens No. 21, 22 and 23 were vacuum annealed at 600 C for 1 hour after rolling.

Specimens No. 31, 32 and 33 were vacuum annealed at 600°C for 3 hours after rolling.

Specimens No. 1, 2 and 3 were made from 3 x 9/16 x 1/8 inch as-rolled uranium sheet and copper as above.

Discussion of Results:

Determination of the effect, if any, of vacuum annealing on the strength of the bond of silver plating to uranium was not achieved by the test because the plating adherence proved stronger than the mercury amalgam bond.

The bond strengths of these specimens (see Table II) were much greater than were indicated by the previous tests (see Table I). Explanation may lie in the possibilities that the bonds in the previous tests were weakened by machining; pressure distribution in the previous tests was uneven and contact poorer because the thin copper warped when machined, and because there were four specimens in each vise. The type of specimens used previously curl when pulled in the tensile tests and are thus subject to a different stress distribution. Since values of 4000-4500 psi were obtained despite oxidation undermining of about 20% of the plating on the uranium, values as high as 6000 psi might be expected since the strength increases with time and temperature of bonding.

The low strengths of specimens 1, 2, and 3 might be attributed to the facts that the surface of the rolled uranium was not as flat as the machined uranium and the thickness of the uranium was not uniform, thereby preventing pressure of the vise on part of the area.

Three specimens similar to Nos. 11, 12, and 13 were fractured by bending the copper away from the uranium. The fracture was of the mercury-silver bond.

Fig. 11 is a cross section of the mercury amalgam bond between silver plated uranium and silver heated in a vise at 350°C for 4 hours.

E. Mercury Bonding of Silver Plated Copper.

Using a hydraulic press with electrically heaten platens, the effect of time of heating on the bond strength of silver plated copper bonded to silver plated copper by mercury amalgamation was investigated.

Procedure:

(1) Sixteen 3½ x 3 x 3/16 inch rolled copper plate specimens were plated with 0.003 inches of silver.

(2) 3½ x 3 inch surfaces were amalgamated with mercury and pressed together under 6,100 psi at 350 C for times of ½, 1, 1½, and 2 hours.

(3) Two bond strength specimens were cut from each of the eight sandwiches as shown in Fig. 10.

(4) Specimens were pulled at a rate of 0.05 inches per minute. The results are given in Table III and plotted in Fig. 11.

Discussion of Results:

Fig. 11 shows an increase in strength with bonding time from $\frac{1}{2}$ hour to 2 hours when the bonds were made at 350°C and 6,100 psi pressure.

[redacted]

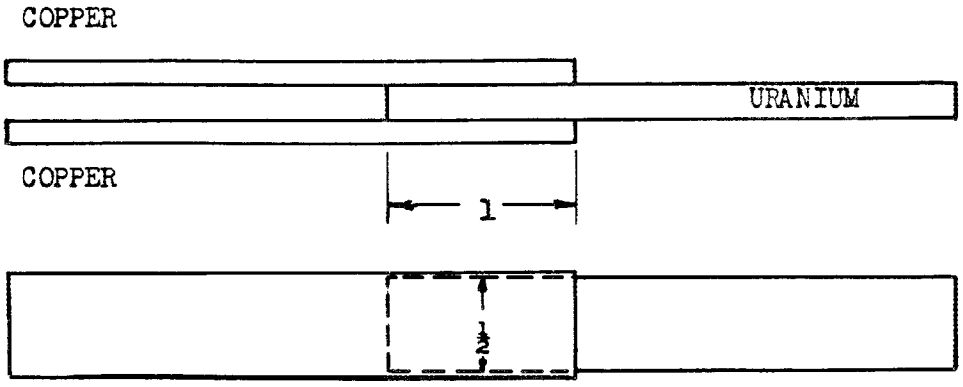


Figure 6

Bond Strength Specimens

Silver Plated Copper to Silver Plated Uranium by Mercury Amalgamation Pressure Bonding

Dwg. No. 10096
Secret

[redacted]

TABLE II

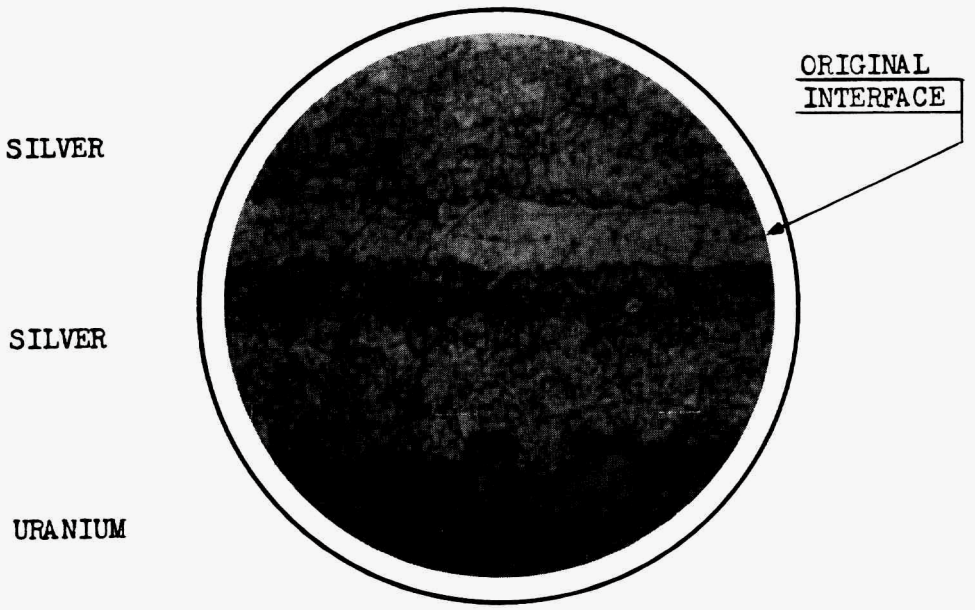
BOND STRENGTH TESTS

Silver Plated Copper Bonded to Silver Plated Uranium by Mercury Amalgamation
Temperature - 350°C Time - 4 hours

Specimen	Vacuum Annealing of Uranium	Maximum Load Pounds or psi[1]	Failure Location	Remarks
11	None	4175	40% Ag-U *2 50% Ag-Ag 10% Ag-Cu	Both bonds failed
12	None	2385	Ag-Ag	One bond failed.
13	None	2620	20% Ag-U 80% Ag-Ag	Both bonds failed
21	1 hour at 600 C	3425	20% Ag-U 80% Ag-Ag	Second bond faile at 1950 pounds.
22	1 hour at 600°C	4385	20% Ag-U 80% Ag-Ag	Both bonds failed
23	1 hour at 600°C	4455		Copper broke befo bond
31	3 hours at 600°C	4400	20% Ag-U 80% Ag-Ag	Both bonds failed
32	3 hours at 600°C	4435	15% Ag-U 85% Ag-Ag	Both bonds failed
33	3 hours at 600°C	4295	15% Ag-U 85% Ag-Ag	Both bonds failed
1	None	3000	Ag-Ag	Second bond faile at 1300 pounds.
2	None	3650	Ag-Ag	Second bond faile at 1810 pounds.
3	None	1835	Ag-Ag	Second bond faile at 1000 pounds.

[1]Total area was one square in in each case.

*[2]All Ag-U failures were around edges and revealed heavy oxidation of uranium.



Y-1954

Mag.-400X Etch - $(NH_4)_2S_2O_8$ + KCN

Figure 9

Silver plated uranium bonded to silver by mercury amalgamation bonding

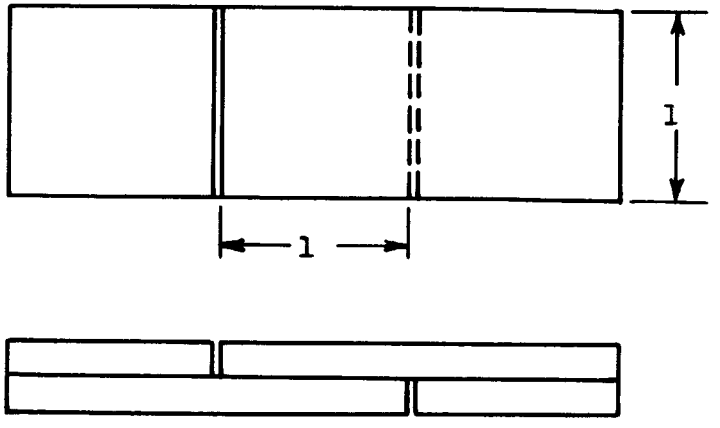


Figure 10

Bond Strength Specimens

Silver Plated Copper Bonded by Mercury Amalgamation

Dwg. No. 10097
Unclassified

TABLE III

Effect of Time on Bond Strength of Silver Plated Copper Joined by Mercury Amalgamation

Bonding Pressure - 6,100 psi Bonding Temperature - 350°C

Specimen No.	Time (hours)	Breaking Stress (psi)	Remarks
1a	½	3780	
b		3500	
2a	½	4020	
b		4450	
3a	1	4420	
b		5000	
4a	1	5900	
b		5730	
5a	1½	5850	Specimen broke outside bond
b		4880	
6a	1½	4240	
b		4470	
7a	2	5380	
b		5200	
8a	2	5970	
b		6220	Specimen broke outside bond

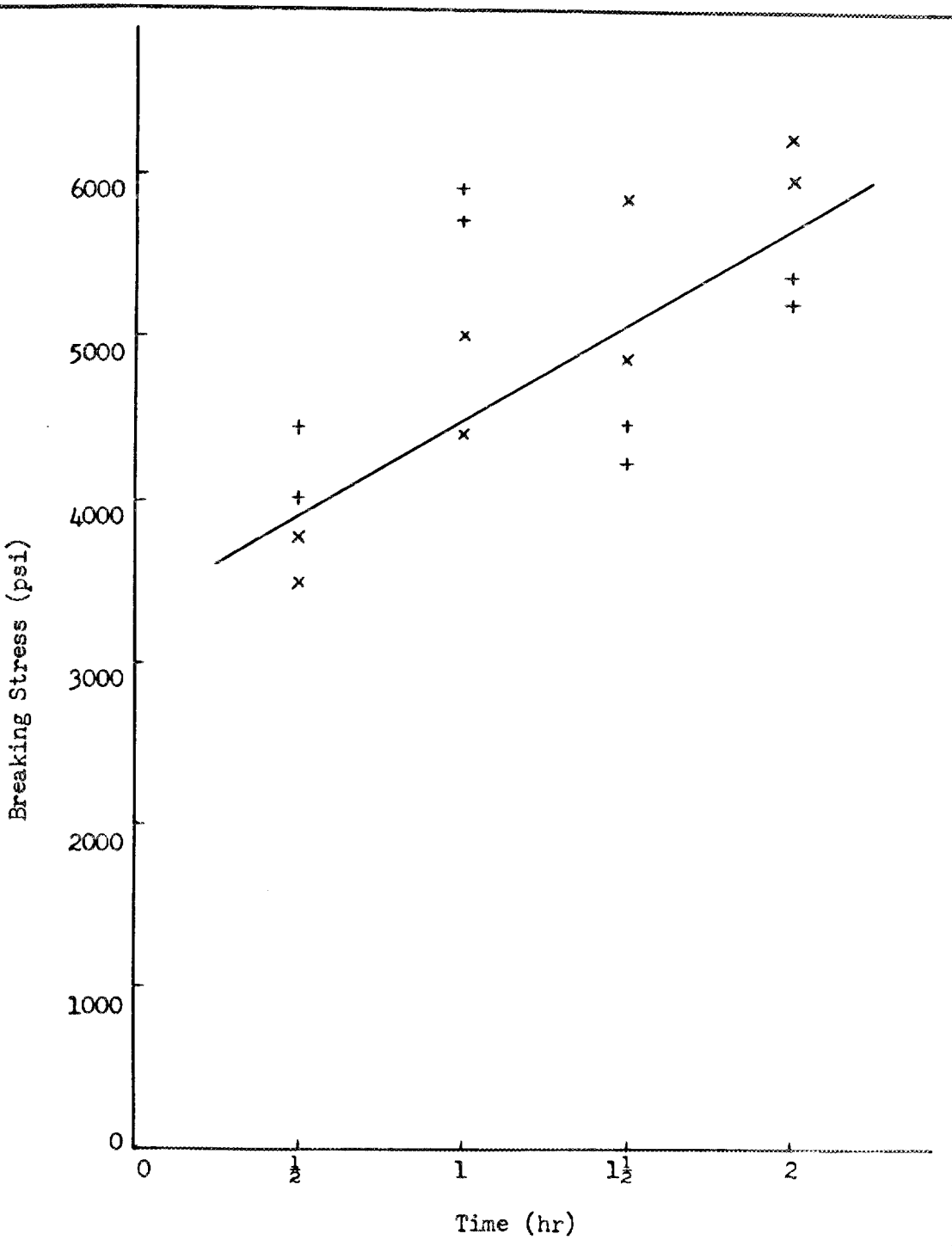


Figure 11 - Increase in bond strength of silver plated copper joined by mercury amalgamation at 350C and 6,100 psi.

Dwg. No. 10098
Unclassified

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(2) L. G. Glasgow, "Report of Visit to BMI," July 17, 1950, CF-50-7-157.

(3) R W. Buzzard and H. E. Cleaves, "The Binary Alloys of Uranium," Journal of Metallurgy and Ceramics, July 1948.

(4) R. E. Eldmann and J. M. Taub, "Fabrication of U^{235} Discs for the General Electric Power Pile," Journal of Metallurgy and Ceramics, May 1949.

(5) CT-2116 A. G. Gray, "The Electroplating of Tuballoy," February 6, 1945.

APPENDIX

Procedure for silver plating uranium:*

(1) Clean anodically in "Anodex" cleaner (6 ounces per gallon) Temperature - 180°F, Current Density - 75 amps/sq. ft.

(2) Rinse in cold water.

(3) Dip for 3 minutes in 1:1 nitric acid (1 part 70% HNO_3 to 1 part water) Room temperature.

(4) Rinse in hot water.

(5) Pickle anodically in phosphoric-hydrochloric acid (50% by weight H_3PO_4 plus 20 ml per liter 37% HCl) Room Temperature, Current Density - 45 amps/sq. ft.

(6) Rinse in cold water 15 to 30 seconds only.

(7) Dip 5 minutes in 1:1 nitric acid.

(8) Rinse in hot water.

(9) Silver plate at 25 amperes per sq. ft.

Bath: 75 grams per liter AgCN
122 grams per liter KCN
225 grams per liter K_2CO_3

Adjust to pH 13 with KOH

Temperature - 113°F

*from Reference (1).