2-5958
12-87958

UCID- 19605

UCID--19605

DESB 004210

PROGRESS REPORT
September-October 1982
Coating Processes Group

J. M. Dini
J. G. Romo

November 19, 1982

This is an informal report intended primarily for internal or limited external distribution. The opinions and conclusions stated are those of the author and may or may not be those of the Laboratory.

Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under Contract W-7405-Eng-48.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

332

2-8342

DISCLAIMER

This report was prepared as part of the work performed by the University of California Lawrence Livermore National Laboratory for the United States Government under contract number W-7400-ENG-48. The report contains certain information which is classified as "Confidential" by the United States Government. This information is being furnished to you for your information only and is not to be distributed outside your organization without the express written permission of the Lawrence Livermore National Laboratory. The information contained herein is not to be used for any purpose other than that for which it was prepared and is not to be used in any way that might result in the disclosure of classified information to unauthorized persons.

November 15, 1982
JWD-82-402

TO: Distribution
FROM: J. W. Dini/J. G. Romo
SUBJECT: Progress Report - September-October 1982
Coating Processes Group

SUMMARY OF CONTENTS

	<u>Page No.</u>
<u>Technical Highlights</u>	2
<u>Weapons Program</u>	
W-84 Ion Plating at Y-12 - A. Copeland	2
W-84 Ion Plating at VPL - A. Copeland	2
W-84 Solid State Bonding - A. Copeland	2
<u>Nuclear Test Program</u>	
Diamond Ace - J. Karcher	3
Tomme Event - C. Holm	3
X-Ray Detector Cathodes for Cabra - C. Bruns	3
X-Ray Detector Foils for Cabra LOS-9 - C. Hawkes	4
Aluminum on Parylene for Cabra Filters - G. Hattery	4
Copper on Parylene Backed Magnesium - G. Hattery	4
Target Foils for Cabra LOS-8 - G. Hattery	4
Target Foils for Cabra LOS-8 - B. Junk	5
Crowdie - C. S. Brooks	5
Pinex Pinhole Development - Z. Briggs	5
<u>Laser Program</u>	
Electropolishing Nova Spatial Filters - H. Patton	5
<u>MFD Program</u>	
RTNS-II - C. M. Logan	5
<u>Miscellaneous</u>	
ME Research - Solid State Bonding Using Electroplating - W. Feng	6
LODTM - Alpha Claddings - S. Patterson	6
Salvaging of Precious Metals - J. Loy	7
Nafion Cell Electrode Coating - G. Haugen	7
Thermocouple Holder Braze - R. Kudo	8
X-Ray Mirror Coatings - G. Sanger	8
NASA X-Ray Mirrors - G. Sanger	8
Micro-Channel Plate Camera - D. Price	8
Laser Flash Tube Failure - K. Whitham	9
Miscellaneous Non-Routine Vacuum Processes Jobs	9
<u>Presentations</u>	9
<u>Publications</u>	10

University of California

☐ Lawrence Livermore
☐ National Laboratory

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

[Handwritten signature]

Technical Highlights

Weapons Program -- We are continuing to support the aluminum ion plating effort for the W-84 both at Y-12 and in-house.

Diamond Ace and Cabra -- Heavy support from VPL both in vacuum engineering activities and fabrication of foils.

Laser Program -- Electropolishing of spatial filters for Nova is on schedule.

ME Research -- This completed program showed that sound solid state joints can be obtained with silver electroplated on a variety of metals.

WEAPONS PROGRAM

W-84 Ion Plating at Y-12 - A. Copeland

We have helped Y-12 complete the process development portion of providing aluminum protective coatings on the W-84 parts. Basically, conventional ion-plating was tailored to suit the unique geometries and material constraints of the assembly. Tests being performed on mock assemblies are showing that the ion-plated aluminum coatings are providing the required protection. Permeability is not detectable, thicknesses are well within tolerance (200 to 1000 microinches), and adherence is quite acceptable. Additional permeability tests are in progress. The next phase of this project is to transfer the process from the Y-12 Development Division to the Y-12 Production Division.

W-84 Ion-Plating at VPL - A. Copeland

In parallel with helping Y-12 satisfy immediate coating needs, we have started a project to develop a more controllable coating process. Our plan is to ion-plate using Magnetron sources instead of conventional resistance heated filament sources. This project has been addressed at a much lower priority level than the immediate Y-12 needs. The hardware is 50% complete and the initial runs are tentatively scheduled for early December 1982.

W-84 Solid State Bonding - A. Copeland

We have electrodeposited silver on 24 test specimens of various metals utilizing procedures developed on our recently completed ME Research Program on solid state bonding. Bob Rosen will have these units welded into containment cans for autoclave solid state bonding and will then test them to failure.

Nuclear Test Program

Diamond Ace - J. Karcher

The full time support of the Diamond Ace experiment vacuum systems was concluded in September.

Due to leakage in the vacuum system, the pressure at shot time was on the order of 50 to 60 microns (5 to 6×10^{-2} Torr) well above the design pressures of 10^{-6} Torr which were proven in July and August.

Even at the higher pressures, some 80% of the experimental data was recovered and the event is considered a successful physics experiment.

Component tests are being conducted in order to determine possible failure modes which may have not been caught in previous testing. Part time support of this work is expected to continue through mid December.

Tomme Event - C. Holm

Design work has begun on another tunnel shot similar to the Diamond Ace event. The experiment has a scheduled FDR in August 1983.

Direct support of the Nuclear Test Engineering Division (NTED) for the necessary vacuum engineering will come from VPL in conjunction with support from EG&G, San Ramon. A requirement for full-time support is expected to extend from January '83 through the balance of the shot.

X-Ray Detector Cathodes for Cabra - C. Bruns

We evaporated cesium iodide (CsI) on aluminum X-ray detector cathodes. The side requiring the CsI coating was diamond machined, making handling more difficult. The CsI coating had to be kept protected from moisture. CsI absorbs moisture quickly and the coating breaks down within minutes of exposure to room air. We installed a glove bag enclosing the vacuum system and kept it filled with argon under positive pressure. The shipping container was also inside the argon-filled bag. Parts and the vacuum system were therefore constantly kept in a dry argon environment. We recommend that holding fixtures and all related parts that are to be used in such an environment be made with the consideration that manipulation is difficult when gloves must be used under positive pressure. The glove bag technique improved our ability to handle parts requiring a moisture-free environment.

X-Ray Detector Foils for Cabra LOS-9 - C. Hawkes

We coated Parylene and Formvar foils on both sides with a nucleating layer of copper and then zinc as an overcoat. The foils had to be pinhole free. The zinc would not adhere without a copper nucleating layer. This meant that four complete coater cycles (one each for copper and zinc and this repeated for each side) and two different vacuum systems would be required to produce a part. We devised a set-up and switched to a multi-source vacuum system in which an electron beam gun was used. This allowed both copper and zinc to be deposited in one coater cycle, in the same coater, cutting down coating time considerably. Techniques for evaporating zinc on fragile Parylene and Formvar foils were improved. More data on the diffusion of copper into zinc under these conditions needs to be gathered.

Aluminum on Parylene for Cabra Filters - G. Hattery

We deposited aluminum onto Parylene by electron beam evaporation. The foils were attached to holders for use as filters for Cabra. One criteria for the foils was that they be pinhole free after coating. We took elaborate steps to try to eliminate pinholes. One technique that helped was depositing half the coating on each side of the foils. This technique, together with blowing off the foils lightly with deionized air just prior to closing the coating chamber succeeded in limiting, but not completely eliminating, the pinholes. The foils did satisfy the requestors needs.

Copper on Parylene Backed Magnesium - G. Hattery

We deposited copper onto existing Parylene backed magnesium foils by electron beam gun evaporation. The purpose of the copper was to cover pinhole defects in the foils so that they could be utilized in the Cabra shot. The foils had been fabricated by an outside vendor (Lebow) but failed to meet specified light-tight requirements. The copper deposition did not plug the pinholes through both the magnesium and the Parylene. Pinholes through the magnesium, but not the Parylene, were sealed. Therefore, there was an improvement, but not enough to meet the requirements. A clean room environment is necessary to reliably fabricate pinhole free coatings and foils.

Target Foils for Cabra LOS-8 - G. Hattery

We coated Parylene foils on both sides with zinc on a nucleating layer of copper. These parts replaced foils that had used aluminum as a nucleating layer and had delaminated. The foils had to be pinhole free. We experienced no delamination. The foils did tend to become loose (i.e., not taut) during the coating process. Coating rate changes affected the foil tautness, but no taut foils were produced using the electron beam gun. We produced taut foils using a resistance heated source, but the only available system could only evaporate one type of material without time consuming adjustments between runs. It was therefore not used to produce the parts. The foils produced by electron beam gun were accepted for the application.

Target Foils for Cabra LOS-8 - B. Junk

We evaporated zinc on a nucleating layer of aluminum, which was evaporated onto Parylene foils. The foils had to be pinhole free. We therefore evaporated zinc on both sides of the Parylene so that any pinholes in the zinc coatings would be offset. We had coated Parylene with zinc using a copper nucleating layer and expected the aluminum to behave similarly. There was no apparent difference until after the finished parts were delivered. The parts then delaminated. Further tests comparing the nucleation of zinc on copper with zinc on aluminum indicated that nucleation of zinc on aluminum was very poor, compared to copper. Coatings of zinc on aluminum may be practical in thicknesses greater than 700 nm.

Crowdie - C. S. Brooks

Electroformed parts requiring three different coatings are required by January 2. These parts are especially challenging since they require thicker coatings than we've ever done and a deposit which we have never produced to this requirement. The first stage of the electroforming process has now been completed.

Pinex Pinhole Development - Z. Briggs

We have produced 11 pinholes for Branco with 2 mil and 4 mil diameter holes by 1 inch long and are currently electroforming 8 units in the 6 and 8 mil size. Our next challenge is to produce 1/2 mil diameter pinholes that are one-half inch long.

Laser Program

Electropolishing Nova Spatial Filters - H. Patton

The total number of spatial filters that need to be electropolished include 72 for Nova and 18 for the two beam lines that will be assembled in France. We have finished 18 of these parts and estimate that it will take until February 1983 to finish the remainder.

MFD Program

RTNS-II - C. M. Logan

Because of oxidation of the tritide on several 23 cm targets, there was a need to process via the wax electroforming method 20 new targets on a short time frame schedule. This was done and some of these targets are already at Y-12 for tritide loading.

A proposal which suggests various electrodeposition schemes for providing a substrate material capable of meeting the property requirements of RTNS-II (e.g., a yield strength of at least 50,000 psi after 4 hours at 400C and an electrical conductivity of at least 80% that of pure copper) has been funded by the RTNS program. Three approaches will be evaluated: (1) electrodeposition of a dispersion hardened copper, (2) electrodeposition of hard copper pyrophosphate, and (3) electrodeposition of copper alloys. We are just beginning to start working on this program.

Miscellaneous

ME Research - Solid State Bonding Using Electroplating - W. Feng

Stainless steel and some other metals were solid state bonded with the aid of electroplated silver. Sound, strong joints were obtained with 1/2 inch diameter cylindrical butt specimens under a variety of bonding conditions. The procedures used for preparing the substrates for plating were first characterized by using ring shear tests which provided quantitative information on bond adhesion. For the bonding studies, a full factorial experiment with two levels and four variables (2^4) was run with stainless steel. The four variables included in the study were: time (1 hr. and 4 hr.), temperature (300C and 600C), bonding pressure (10,000 psi and 30,000 psi), and silver plating thickness (1 mil and 6 mil). With stainless steel, the mean bond strength was 47,800 psi with failure occurring within the silver electrodeposit. Time was a non-significant variable, while thickness, pressure and temperature were all significant as were the interactions of temperature with pressure and temperature with thickness. Although full factorial experiments were not run with other materials, enough data were obtained to reveal that time was insignificant for these materials, temperature was very significant and thickness and load relatively insignificant. A final report has been written on this project and is presently going through the approval process.

LOUTM - Alpha Claddings - S. Patterson

A. Hard Copper

We are investigating deposition of a hard copper deposit for the Alpha program. The goal is to produce a deposit that will be hard and yet not change properties, e.g., lose hardness and strength as a function of time. Preliminary work with a pyrophosphate copper solution containing a proprietary addition agent (PY-61-H) has shown promising results. Data presented in Table I show that deposits produced in solutions containing 500% and greater than vendor recommended amounts of the additive were stronger and retained their strength even after exposure to 250C for 4 hours.

B. Plating on Molybdenum

Two procedures for plating on molybdenum were evaluated:

1. Bell Labs Process - the key steps in this method include two firing operations in dry hydrogen at 1000C.
2. Hughes Aircraft Process - the key steps in this method include a multitude of firing steps at 800C.

Ring shear tests were used to obtain quantitative data for gold plated on molybdenum which was treated by the above procedures. Results showed consistently higher values for the specimens prepared via the Bell Labs method, e.g., bond strengths of 18,000 psi were consistently obtained and failure occurred entirely within the gold deposit. Specimens treated by the Hughes Aircraft process exhibited an average failure of 11,500 psi with failure often involving separation at the molybdenum-gold interface.

Table 1

Ultimate Tensile Strength of Pyrophosphate Copper
as a Function of Brightener Concentration

<u>Brightener Concentration* (%)</u>	<u>Tensile Strength (K psi)</u>		
	<u>As-Deposited</u>	<u>2 Hrs at 250C</u>	<u>4 Hrs at 250C</u>
0	64	39.5	44
100	79.5	47.5	41.5
200	81.5	46	51.5
300	83	61	56
400	80	60	62
500	89	79	82
750	92.5	83.5	89
1000	83	83.5	91

*PY-61-H, M&T Corp., Rahway, NJ

Miscellaneous

Salvaging of Precious Metals - J. Loy

Over the past five years or so, Materials Management has built up a backlog of precious metal plated parts that are scrap. These include printed circuit boards, reflectors, targets and a variety of other parts. We salvage the precious metal coatings on these parts, and to date this year have recovered 60 troy ounces of gold.

Nafion Cell Electrode Coating - G. Haugen

We electron beam evaporated gold onto both sides of two nickel electrodes. The Nafion is sandwiched between the two electrodes to maintain contact during the swelling which occurs due to the Nafion absorbing water. The initial request was for us to deposit nickel through a grid pattern directly onto Nafion. During the requester's experiment, the nickel coating would peel off due to the distortion caused by the Nafion absorbing water. Using the gold coated nickel electrodes eliminated the problem of losing constant current flow to the Nafion which is used to electrolyze water into hydrogen and oxygen.

Thermocouple Holder Braze - R. Kudo

We were asked to vacuum braze a stainless steel cap to the end of an electrolytic tough pitch (ETP) copper tube for use as a thermocouple holder. ETP copper is not desirable for vacuum brazing because it is difficult to reliably achieve leak tight joints. Another special consideration was that we were requested to use a braze alloy that did not contain nickel. This forced us to use a less desirable alloy, Silcoro 75, which resulted in a vacuum leak at the joint. Numerous attempts to seal the leak by remelting the alloy failed. As a last resort we used Cusil, a lower temperature alloy, and succeeded in plugging the leak. Cusil is not normally used to braze to SST without first nickel plating. However, in this case it was able to wet to the Silcoro 75 and close the leak.

X-Ray Mirror Coatings - G. Sanger

In conjunction with Optics we are developing a coating process to produce X-ray mirrors consisting of very thin alternating-layers of two dissimilar metals on glass substrates. Initially we have produced three specimens, by Magnetron sputtering, each having a total of 101 layers. The thickness of each layer is 3 nm on one specimen, 1.5 nm on another specimen, and only 0.5 nm (5Å) on another. The Optics Group will be evaluating the results. A shutter mechanism was devised to precisely meter the deposition intervals. The EE Machine Control Group produced a controller that provides intervals of one-to-99 seconds and a repetition rate of one-to-999 seconds. The use of two (50 mm diameter) Magnetron guns instead of two electron-beam guns has resulted in a substantial savings of both time and money.

NASA X-Ray Mirrors - G. Sanger

We were asked to deposit adherent coatings of copper, with a chrome interface, on quartz parts for the subsequent fabrication of X-ray mirrors. The coatings serve as a base for electroplating a 3 mil thick coating of copper which is then diamond turned to a mirror finish. For best adhesion, the parts were heated to 300°C during the PVD process. We saved time by fabricating a special fixture that permitted coating up to three parts each run.

Micro-Channel Plate Camera - D. Price

We have developed a technique for joining Al_2O_3 to a piece of SnO_2 coated fiber optics glass. A test braze was completed to prove the feasibility of making such a joint for the fabrication of a micro-channel plate camera. The problem was to create a good joint between two ceramic materials at a low enough temperature to not break the glass. The braze alloy Eutectic 157 was chosen because of its low melting temperature of approximately 260°C. Initial tests showed that of the materials considered for an interface, this alloy would wet copper best. Both test pieces were vapor plated by the requestor with a 20 nm base layer of chrome (to promote adhesion) and a 200 nm layer of copper. The braze appears to have worked well, with braze alloy wetting both surfaces. The joint is currently being tested for vacuum tightness.

Laser Flash Tube Failure - K. Whitham

We were asked to assist in (1) determining the reason for failures in laser flash lamps, and (2) recommending a remedy. The commercially made lamps have been failing at various stages of their life by developing breaks at the tungsten to glass seal. At a meeting, involving the requestors, members of VPL and the glass lab, we offered some ideas that we feel should reduce the breakage problem substantially. The ideas involved modifying the lamps. This would be desirable because a large number of the lamps already exist.

Miscellaneous Non-Routine Vacuum Processes Jobs

Tested contents of Diamond-Ace gas sample bottles - W. Gregory.
Induction and radiant heat brazed streak camera components - J. Bushman.
Evaporated boron foils for Cabra LOS-9 - G. Hattery
Determined outgassing characteristics of RF gasket - J. Karcher.
Tested Diamond-Ace vacuum gauges - J. Kroger
Provided SEM vacuum system maintenance - C. Witherell.
Determined outgassing characteristics of epoxy coated aluminum - J. Karcher.
Furnace tested CRI-012 part - C. Brooks.
Induction brazed Advac to SST fittings - C. Gillium.

Presentations

1. W. K. Kelley, W. C. Cowden, and G. E. Overturf, "Electroforming Cylindrical Parts with Precision Small Inside Diameter Pinholes", ICMOC/SUBJOG-12A, September, Rocky Flats.
2. W. K. Kelley, "Use of Electrodeposition to Provide Coatings for Solid State Bonding", same meeting as 1 above.
3. T. G. Beat, G. E. Griggs, "Quantitative Adhesion Data for Two Energetic PVD Processes", Same meeting as 1 above.
4. T. G. Beat, J. G. Romo, G. Griggs, D. Berger, R. Reiss, and T. Brown, "Fabrication of Foils for Caboc", same meeting as 1 above.
5. J. G. Rittmann, "LLNL/Materials Fabrication Division Specialty Facilities/Coatings Overview" same meeting as 1 above.
6. W. K. Kelley, G. E. Overturf, J. W. Dini and D. P. Atkinson, "Joining a Large Brass Ring to Stainless Steel with the Aid of Electroplating", Electrochemical Society I-62 Meeting, Detroit, Mich., Oct.
7. J. W. Dini and H. R. Johnson, "Plating on Titanium and Zirconium", ASTM Conference on Titanium and Zirconium in Industrial Applications, New Orleans, Sept. 21, 1982.

8. J. W. Dini, "Influence of Impurities on the Properties of Nickel Electrodeposits", Seattle Branch, American Electroplaters' Society, Oct. 13, 1982.
9. J. W. Dini, "Plating on Difficult to Plate Metals", Portland Branch, American Electroplaters' Society, Oct. 14, 1982.

Publications

1. D. D. Berger, "Calcium Fluoride Window Mounting", Review of Scientific Instruments, 53 (10), 1619 (1982).
2. J. W. Dini, "Electrodeposition - A Viable Coating Alternative", Thin Solid Films, 95, 123-129 (1982).
3. J. W. Dini and H. R. Johnson, "Plating on Titanium and Zirconium", UCRL-87111, Sept. 1982.
4. J. W. Dini, F. B. Waldrop and R. W. Reno, "Surface Preparation and Plating", UCRL-88199, Oct. 1982.

J. W. Dini
Coating Processes Group
Materials Fabrication Division

J. G. Romo
Coating Processes Group
Materials Fabrication Division

JWD:JGR:jm
1956R