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TARGETS BY ELECTROPLATING

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MASTER

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METAL COATINGS FOR LASER FUSION TARGETS BY ELECTROPLATING*

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ABSTRACT

Metal coated laser fusion targets must be dense, uniform, spherically symmetric to within a few percent of their diameters and smooth to better than a few tenths of a micron. Electroplating offers some unique advantages including low temperature deposition, a wide choice of elements and substantial industrial plating technology. We have evaluated electroless and electroplating systems for gold and copper, identified the factors responsible for small grain size, and plated glass microspheres with both metals to achieve smooth surfaces and highly symmetric coatings. We have developed plating cells which sustain the microspheres in continuous random motion during plating. We have established techniques for deposition of the initial conductive adherent layer on the glass microsphere surface. Coatings as thick as 15 μm have been made. The equipment is simple, relatively inexpensive and may be adopted for high volume production of laser fusion targets.

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Introduction

The difficulty of metal coating laser fusion targets is compounded by the small size of the targets and by the severe material constraints which must be met by the coated targets. Typically targets are formed from hollow glass microspheres which may be from 70 to 200 μm in diameter. The microspheres are filled with high pressures of a deuterium tritium gas fuel mixture. Metal coatings applied to these microspheres must be spherically symmetric to within a few percent of their diameters smooth to 0.1 μm or better and be full bulk density material.

Three major problems were obstacles to electroplating laser fusion targets. It was first necessary to metallize (strike) the glass microspheres to provide an electrically conductive, platable surface. We have used both wet chemical and physical vapor deposition¹ processes as techniques to metallize glass microspheres. The superior surface finish of thin sputtered metal coatings has made these sputtered Pt coatings the present best strike choice.

The second major impediment to metal coating targets is their fragility. The metallized glass substrate is susceptible to damage or fracture from impact, thermal shock, or from external stress due to overlying coating layers, or from target transfer operations. Copper and gold are relatively soft metals consequently coatings of these metals are also susceptible to damage from vacuum welding, impact deformations as

well as from imperfections in the coating process itself. Therefore, we had to develop plating cells which sustained the microspheres in gentle random motion against the surface of a suitable cathode during electroplating. We have developed and used two new cell types whose plating action is very gentle. We were unable to use designs similar to Mayer's previously published design² because damage to the substrate or coating occurred using a cell of that type.

Finally we chose the plating solution and plating process parameters to optimize the material properties of the metal coating deposited using our plating cells. Small grain size, freedom from porosity and low mechanical stress were the material properties we sought. Practical metal coatings for laser fusion targets must also be strong enough to withstand the assembly and analytical processes and stable enough to accept overcoating of ablaters or diagnostic layers.

II. Experimental

The initial metallization of the microspheres was carried out by magnetron sputtering. These sputtered platinum coatings were deposited from 1000 to 1500 Å thick on DT filled microspheres with the same equipment and techniques as previously reported.¹ Surface smoothness was very good, with irregularities of ~10 nm arising primarily from imperfections in the glass substrate. Deposition of these thin platinum coatings required less than 10 minutes of sputtering. No loss of DT fill

from the microspheres was observed due to this short exposure to the sputtering plasma.

Figures 1 and 2 show respectively the vibrating cathode cell and the tapered cathode cells which we have developed. The vibrating cathode cell uses mechanical vibration of the cathode screen coupled with pulsed variation in the rate of solution flow from the cell to provide random motion of the initially boyant microspheres during plating. The tapered cathode cell shown in Figure 2 utilizes a balance between solution flow, viscous forces and bouyant forces acting on the microspheres to provide a rolling motion along the walls of the cathode as the microspheres rise from the lower part of the cell. Near the neck at the top of the cell the bouyant microspheres are swept off the walls and down to near the bottom of the cell only to rise again and repeat the plating cycle. We have observed motion of the microspheres in a glass version of this cell and infer from such observations that the action during plating is as described.

We have chosen commercially available pyrophosphate solutions for copper plating and commercial sulphite gold solutions for gold plating. Although the solutions, the plating cells and material properties of the plated deposit are coupled parameters both literature descriptions of plating results and our experience support the choice of pyrophosphate and sulphite systems in general and the commercial solutions in particular.^{3,4} We have sought to make improvements in the material

properties of the electroplated coatings using the results from the unmodified commercial solution as benchmarks.

For copper plating a commercially available pyrophosphate solution was used both at (21°C) and at 45°C (M & T Chemicals-Unichrome). The temperature of the solutions was monitored by sheathed thermocouples placed 2 cm from the exit of the plating cell. The heat capacity of the solution was utilized to provide a constant temperature at the plating cell. The solution was maintained per manufacturers specifications for plating carried out at 45°C. Plating at 21°C, we found a 10% increase in solution concentration was desirable. An additional 10% increase in the pyrophosphate component enhanced the copper deposition rate at 21°C and reduced the grain size and the internal stress in the copper coating. The pH was maintained at 8.4 with analytical reagent grade ammonium hydroxide. The Gold plating solution we used was BDT-510 marketed by Sel Rex Division of Oxy Metal Industries. The solution was used at room temperature, recommended concentrations and pH, but without the addition of the brightener. Adjustments in component concentrations were made either with analytical reagent grade materials or in the case of proprietary solutions with reagents purchased from Sel Rex.

We investigated both conventional DC plating and pulsed DC plating. Although both plating cells were operated with DC power supplies and pulsed DC supplies, only the copper solution was used in pulsed DC experiments. We found, in agreement with some previous workers, that

gold deposits from sulphite gold solutions are not improved by the application of pulsed DC.⁵ During conventional DC copper plating currents of 4 and 8 mA were drawn respectively by the tapered cathode and vibrating cathode cells from constant current power supplies. Gold plating was done with 6 or 8 ma DC for the respective cell types. Current densities for both cell types were a factor of 10 less than the density for which autocatalytic behavior has been observed in some sulphite gold systems.⁶ Typically cathode current density was 0.4 ma/cm^2 or less for DC plating of both gold and copper. During copper plating with pulsed DC, a 4.5 volt one millisecond square DC pulse was repeated every 4 milliseconds, to draw fifty millamperes of current.

The pulse generation equipment used for copper plating was based on Tektronix TM500 series modules. Switching was accomplished with a LLNL designed solid state switch capable of handling currents up to 3 amperes. Rise and fall times at 100 ma current were 50 nanoseconds. Currents and voltages were oscilloscope displayed. A Tektronix Hall effect probe was used to sense the currents to the cells.

The mechanical vibration applied to the cathode in the vibrating cathode cell was applied through a Ling force transducer driven with an amplified sine wave pulse. A variety of square waves, ramps and sine waves were amplified and used to drive the Ling transducer. Frequencies from .01 cps to 100 cps with amplitudes of from .1 mm to 2 mm were utilized to drive the cathode screen. The drive rod and cell were

equipped with mechanical stops to prevent rupture of the screen. Solution flow from either cell was regulated with a solenoid valve driven by a variable clock. Solution was allowed to flow for 2 seconds and then the valve was closed for 5 seconds in the tapered cathode cell. The valve was closed for 20 seconds in the vibrating cathode cell. These solution pulse cycles were repeated for the duration of the plating. Experiments were also conducted with steady state flow rates of 500 cc/hour for the vibrating cathode cell and 900 cc/hour for the tapered cathode cell.

The water used in washing plated microspheres and in the makeup of solutions was filtered low conductivity water which was distilled in a tin lined still. The aqueous wash procedure following microsphere plating included three 60° water washes followed by an air drying step. Analysis steps carried out on coated microspheres included surface examinations by SEM, radiography for thickness measurements and Auger examination of relevant surface defects.

III. Results and Discussion

Both the copper and gold are being electroplated under static or near static conditions. This fact severely limits the currents which can be applied to the cells. Rates are therefore relatively low when compared with larger scale plating processes. Typically, for both copper and gold, the coating rate is about 12 m per hour. Since the coating rate

for both cells is dependent on the density of the microspheres and their cathode contact time, we find a distribution in coating thickness within a batch of microspheres. In order to copper plate in excess of $7\text{ }\mu\text{m}$ or gold plate thicker than $4\text{ }\mu\text{m}$ (on $140\text{ }\mu\text{m}$ diameter microspheres) the currents supplied to the vibrating cathode or tapered cathode cell must be reduced progressively as the coating proceeds. This reduction is necessary because the mass of the coating becomes significant when compared with the mass of the microsphere. Mayer's design is effectively density independent because the rapidly flowing plating solution forces the microspheres into contact, consequently his plating rates are a factor of 5 higher than ours.²

Figure 3 displays the reductions in grain size of the copper deposited on $140\text{ }\mu\text{m}$ diameter microspheres we have obtained. Figure 3A shows typical room temperature results using manufacturers recommended solution parameters. The grain shown in Figure 3A could no doubt be reduced by using the solution at the manufacturers recommended operating temperature of from 45 to 55°C , however, concern for potential loss of DT fill or breakage of filled microspheres lead us to investigate the range of improvements available at room temperature. Figure 3B demonstrates that proper choice of current density and solution parameters markedly influence coating morphology even at room temperature. Very low current densities increased grain size as did very high current densities. A fifty percent variation from the optimum currents for any cell or solution combination produced little change in

coating morphology. Changes in excess of $\pm 50\%$ of the optimum current density were observed to increase the grain size in the coating. A factor of 3 increase from optimum in DC currents seriously eroded solution stability.

The grain size observed in copper coatings deposited at room temperature was very sensitive to the solution operating parameters. A pH change of 0.25 units caused a 30% increase in grain size. A 10% increase in pyrophosphate concentration resulted in a 30% decrease in grain size at a constant solution pH. An overall 10% increase in initial solution concentration reduced the grain size another $\sim 30\%$. Figure 3C shows the results of the combined effect of proper solution concentrations, pulsed solution flow from the cell and best current density combined with the DC pulse plating technique. Figure 4 shows the typical surface of a copper microsphere plated with copper under the present optimum combination of operating conditions.

The morphology of gold electroplated from BDT-510, in contrast to copper, was nearly insensitive to solution variations. The grain size observed in gold coatings was always smaller than that observed in equivalently deposited copper. Under the best plating conditions, gold grains are perhaps 40 to 60 Å in size. Figure 5 shows three views of a 2 μm thick gold plated glass microsphere.

For both gold and copper the variation in coating surface quality with amplitude and frequency of vibration for the vibrating cathode cell was not significant. No significant difference was found among the various pulse types because the inertia of the system limited its response to drive variations. What did vary with vibration conditions was the number of microspheres which became plated to the cathode. When vibration of the cathode was optimum the pulsed variation in solution flow allowed a 95% recovery of the initially loaded microspheres. Without pulsed solution operation 80% of the loaded microspheres were recovered from the vibrating cathode cell. The 20% remaining were plated to the cathode. The effect of solution pulse and recovery amounts for the tapered cathode cell were virtually identical to those results described above for the vibrating cathode cell.

Mayer, et al, have successfully deposited electroless nickel from commercially available plating baths.² Although electroless nickel plating baths are highly developed, widely used methods of nickel deposition, nickel is not an element of much interest in current target designs.⁷ Unfortunately, we found gold and copper electroless plating solutions do not deposit as smooth, as uniform, or as dense a coating as nickel plating systems do. We investigated electroless gold systems using BH_4 as a reducing agent.⁸ We found that the first gold coatings from BH_4 electroless systems were much less smooth than the first electroplated gold coatings. Figure 6 shows a SEM micrograph of the first microsphere we coated with electroless gold. The inherent

grains are 3 to 5 μm in size. Since electroplating gold and copper showed much more promise, we have terminated further attempts to refine the electroless systems for either metal.

IV. Summary

Figure 7 shows a radiograph of a copper coated ball which has been overcoated with a fluorocarbon ablator.⁹ The example in Figure 7 demonstrates that for this target, in addition to the surface and uniformity properties required of the copper coating itself, this electroplated copper microsphere has withstood assembly and analytical procedures as well as the application of an adherent, uniform and smooth ablator. We expect further progress in surface smoothness and yields will make electroplated metal coatings an indispensable part of laser fusion targets.

We have begun to realize some of the advantages of electroplating technology; rapid deposition at low temperature, relatively simple equipment and deposition of soft metals. We hope to extend the technology to produce cells that operate independent of the density of the substrate and to understand the details of the plating process itself. The possibility of alloy plating raises the prospect of mechanically strong coatings for fuel containers and makes possible a variety of new implosion diagnostics. With greater understanding we can improve and extend electroplating to achieve the high volume production of targets required for an operating ICF fusion power plant.

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Figure Captions

1. Vibrating cathode cell. Solution flows in from the tube at upper right and exits the bottom tube for plating bouyant microspheres. Solution flow may be reversed for plating dense microspheres.
2. Tapered cathode cell. The solution flows from top to bottom for plating bouyant microspheres. Dense microspheres may be plated by inverting the cell and solution inlet. The mode also serves on a flow diverter.
3. Improvements in grain structure of copper electroplated on a 140 glass microsphere from pyrophosphate solution using either vibrating cathode or tapered cathode cells.
 - A. DC electroplated at room temperature
 - B. DC electroplated at optimum current density, room temperature modified solution composition.
 - C. Modified solutio + DC pulse plating.
4. Surface finish of copper coating, DC pulse electroplated on a 140 μm diameter glass microsphere.
5. Smooth uniform gold electroplated gold coating.

6. First microsphere plated with electroless gold coating.
7. Radiograph of electroplated copper microsphere coated with CF ablator. The densitometer trace shows both the outer CF ablator and the inner copper coating to be uniform.

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ELECTROPLATING CELL

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Fig. 1

ELECTROPLATING CELL

Tapered cathode cell

1. Bottom cap
2. Anode
3. Bottom security screen
4. Cathode
5. Anode connection wire
6. Top security screen
7. Top cap

10-05-0880-2548

Fig. 2

Normal

2 μ

Grain refined

2 μ

**Grain refined
and pulse plated**

2 μ

Fig. 3

SURFACE FINISH OF COPPER ELECTROPLATED ON GLASS MICROSPHERE

→ | 2 μ | ←

10-05-0480-1366

Fig. 4

SMOOTH UNIFORM ELECTROPLATED GOLD MICROSPHERE

40 μ → | | ←

20 μ → | | ←

2 μ → | | ←

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Fig. 5

FIRST GLASS MICROSPHERE WITH ELECTROLESS
GOLD COATING

30 μ m

10 μ m

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Fig. 6

**Microradiograph Electroplated copper
with CF ablator**

Densitometer trace