Manufacture and Characterisation of Materials for High Power Laser Targets

A. Hughes¹, D. Haddock¹, W. Nazarov², C. Whyte, M. Tolley¹ and C. Spindloe¹

¹Science and Technology Facilities Council, Rutherford Appleton Laboratory, Harwell Campus, Chilton, Didcot, Oxon, OX11 0QX, UK

Email: aasia.hughes@stfc.ac.uk

Abstract. The Target Fabrication group at the Central Laser Facility (CLF) works to develop and produce novel targets for the high-power laser community. Targets range from simple foils to more complex geometries and with many components and key part of the research is materials development. We describe the recent advances in the CLF in surface preparation for electroplating and the manufacture of thin films. Characterisation of these materials is carried out using Atomic Force Microscope and Scanning Electron Microscopy. These advances have been applied to target components that have been used experimentally on the Vulcan Laser system at the Rutherford Appleton Laboratory.

1. Introduction
The Target Fabrication group in the CLF has been delivering targets to the UK high power laser community for over 40 years with thin films being made from the opening of the CLF in 1978. The Central Laser Facility established a dedicated chemistry laboratory to support its internal program in 2010 and has since then developed the capabilities to manufacture low density polymeric foams using critical point drying techniques, ultra-thin polymer films using dip and spin coating techniques [1, 2] and also electroplating capabilities for micro objects [3]. The focus of the research has been to push the limits of the capabilities to make materials for high power laser experiments through a use of novel techniques. Advances in the characterisation of these materials has given an insight into the microscale features of these target materials and has enabled a greater understanding of the limitations of the processes. This information is fed back to the user community to enable them to design technically possible targets that achieve their scientific aims and this collaboration enables the target fabrication group to focus its future research on the developing needs.

2. Electroplating
We have been developing electroplating capabilities to reduce the need for glue layers for multi-layered and multicomponent targets. Glue layers in such targets can have detrimental effects on the results obtained, in electron transport experiments the addition of a glue layer can complicate the analysis, and in high energy density science experiments where shocks are launched into targets a glue layer can cause reverberations between the layers, again complicating the experiments. We have used flat and structured aluminium substrates to investigate the suitability of this technique for multi-layer target manufacture.

It is important to have good uniformity of electroplated foils when manufacturing them for laser experiments. Target foils are usually <100μm thick. During electroplating nodule formation is common [5]. If nodules are allowed to form during the plating process foil thickness varies across the surface. This result will not meet target criterion. Inconsistencies in foil thickness can have an impact on the laser experiments for which they are produced; therefore one area of focus is to reduce the
number and height of these nodules as successful, uniform plating across flat and structured surfaces also offers the potential reduction of components and assembly processes in target manufacture.

Preliminary work carried out saw copper electroplated directly on to flat aluminium substrates without any prior treatment, other than cleaning the aluminium substrates. This work was done to establish valuable areas of investigation. Figure 1 shows the formation of islands on the aluminium substrate. Figure 2 shows the size variation of the copper islands. These plating results show nodules of up to 154µm proud of the plated surface on a 240µm thick aluminium foil.

There are many factors that can affect the quality of a plated surface, in this case five were chosen; effect of seed layer, pH of plating solution, solution mixing, current applied, and solution temperature. Copper was electroplated onto aluminium foils (dimensions: 0.24x10x40mm). Copper sulphate solution was made in the laboratory from solid copper sulphate crystals, 1 moldm⁻³ solution was made for each test. 240µm thick and 99.0% purity aluminium sheets were cut into 5x1cm strips. All aluminium samples were cleaned with 10% sodium hydroxide solution to remove surface contaminants, following cleaning copper seed layers were deposited using plasma sputter coating on most aluminium substrates. When cleaned and coated, prepared aluminium strips were kept under nitrogen to prevent oxide formation on the surface. Native oxide layers are common and inevitable on aluminium and copper metals exposed to oxygen. [4, 5]

2.1 Seed Layer Effects
An aluminium test strip was plated with no seed layer in 1mol dm⁻³ CuSO₄ solution at 10mA to establish the baseline plating parameters. This test sample did show some copper deposition; however the results gave nodules or islands of copper rather than foils plated over the entire aluminium surface. Nodule formation is investigated in some detail in literature by Guo [6], [7]. Copper had not plated across the whole surface with the edges preferentially plated. Where plating had occurred copper nodules ran along furrows present in the aluminium foil. Results from this run achieved no usable foil or flat area. Nodules formed varied greatly in diameter and height. Figure 2 shows the diameter ranges observed. The modules characterised varied in height, from 3µm-154µm.

A second test piece was coating with a seed layer of copper as described above. Seed layers deposited were 240nm on side A and 180nm on side B. It is clear that this layer greatly enhances the deposition of the copper during the electroplating process. The copper applied showed a marked improvement in uniformity and quality compared to results of plating cleaned aluminium. Figure 3
shows the plating attempt made with a Cu seed layer. Figure 3 also shows the variation in quality across the surface with areas of discoloured and bright copper. Although, the resultant foil is poor in copper quality, the image does show increased foil plating success across the substrate compared to attempts made without seed layers.

Figure 3: Copper plating with seed layer, 10mA applied, 2hours plating time, 1mol dm$^{-3}$ CuSO$_4$ solution concentration.

2.2 Effect of Solution Temperature
The aim of investigating solution temperature was to assess if the thickness of the copper foil plated differed across a temperature range. Three temperatures were investigated; 10°C, 18°C and 40°C. Room temperature of the lab used is 18°C. Plating attempts made at 40°C delaminated to some degree. It was evident that there was some plating success; delamination of the plated foil was deemed unsuccessful. Although this information is useful, this investigation aimed to achieve multi-layered, glue-free targets. Side A (240nm seed layer) delaminated more readily and in larger pieces than side B (180nm seed layer). At 10°C there was little to no change to the foil thickness compared to foils produced at 18°C. The temperature difference may have been too small to cause any change in plating result.

2.3 Current Applied to the Sample
The current applied to the aluminium strips was 5, 10, or 15mA. All samples were processed in a standard set up of 1mol dm$^{-3}$ CuSO$_4$ solution for 2 hours, without continuous mixing, at 18°C. Where 5mA was applied poor results were obtained. In these instances, the copper deposited only as nodules that ranged from 25um-110um in height. The nodules post-plating appeared bright copper coloured. It is unclear the reason for the blue colour change on the aluminium strip observed in figure 4. EDX was carried out to assess the elements identified; only copper and aluminium were identified with no other significant elemental peaks. Figure 4 shows one of the results of this plating attempt. No overall thickness measurements were taken for these samples as there were no areas where usable foil had been plated to obtain consistent measurements.
Figure 4: Image of 5mA no mixing, nodules can be seen across the surface. No foil success.

Where 10mA was applied again poor quality foils were achieved, this is shown in figure 4. Copper plating achieved on side A (240nm seed) gave thicknesses of 18-20um, side B gave foil thicknesses of only 6-8um. It is unclear the reason of thickness variation on sides A and B. The thicknesses vary throughout the investigation and there is no obvious preferred seed layer thickness at this stage. The roughness (Ra) was measured using white light interferometry to be 3.8um on side A and 1.8um on side B. Discolouration on the surface was observed along the width of the sample, following the orientation of the grooves found in the aluminium pre-processing.

Figure 5: 10mA applied, solution was not continuously mixed. Copper thickness varies greatly across entire surface.

Where 15mA was applied foils of copper were achieved. These samples gave thicknesses of 0-28.8um on side A. Side B was uneven giving coating of 0um-17um. The thickness of copper across the surface was arbitrary. Both sides showed areas where plating had not occurred. Although copper had plated onto the aluminium surface, large areas of thick, nodular, dull copper and areas of thinner, flatter, brighter copper were observed. The thicker areas appeared darker in colour, much like the foils achieved at 10mA. The thickness range across the aluminium surface does not meet with the aim of the investigation. Results obtained suggest these parameters should not be used for the purpose of producing thin copper foils.

EDX analysis was carried out on the discoloured samples, significant oxygen peaks were observed suggesting the copper had oxidised. Figures 6 and 7 show images taken with a scanning electron microscope (SEM) of samples plated at 5mA (fig. 4) and 10mA (fig. 5). Although at 10mA plating
had showed greater success than at 5mA, the surface roughness observed implies a greater copper surface area exposed to air, thus increasing the probability of oxide formation.

2.4 Continuously Mixing the Solution
Initial tests carried out without mixing the plating solution gave inconsistent foils and nodules on the plated areas of the substrate. For these tests all other factors remained constant. Current applied was 5, 10, or 15mA in a 1mol dm$^{-3}$ CuSO$_4$, for 2 hours, at 18°C. All test strips showed a marked improvement in uniformity of thickness across the surface when the solutions where continuously mixed. Dull areas remain, and this is where the copper was rougher compared to the appearance of brighter foil sections, foil roughness detailed in section 2.6. Thicknesses achieved can be seen in table 1. These foils adhered well to the aluminium surface making them useful for multilayered target manufacture.

| Current applied | Solution Mixed - Thickness side A (µm) | Solution Mixed - Thickness side B (µm) | Not Mixed - Thickness side A (µm) | Not Mixed - Thickness side B (µm) |
|-----------------|----------------------------------------|----------------------------------------|----------------------------------|----------------------------------|
| 5               | 5                                      | 14-15                                  | N/A too many nodules             | N/A too many nodules             |
| 10              | 20-21                                  | 7-8                                    | 18-20                            | 6-8                              |
| 15              | 9-10                                   | 10                                     | 0-28                             | 0-17                             |

Table 1: Copper foil thickness using white light interferometry.

2.5 Variation of the pH of Solution
To reduce the pH of the solution sulphuric acid (10ml H$_2$SO$_4$ at 50%) was added to the copper sulphate. All other conditions remained the constant as in the previous tests the current applied was 5, 10, or 15mA in a 1mol dm$^{-3}$ CuSO$_4$ solution, for 2 hours, at 18°C. Initial plating attempts made without solution mixing produced nodular foils and therefore due to the foil success achieved by continuous mixing as detailed in section 2.4 this factor was kept constant.

The resulting foils across all currents applied were flatter, appeared bright copper in colour and plated more evenly over the entire substrate. However these foils were easily removed from the
aluminium substrate by peeling. Though useful, our aim was to achieve copper suitably adhered to the substrate to produce layered targets. Figure 8 shows the flatness achieved at 5mA with sulphuric acid. It was observed that 5mA give the best quality foils and this is due to the slower plating rate at this current density. Figure 9 shows the comparison of foils with and without sulphuric acid added to the solution. Table 2 show the variation in thickness achieved under these conditions. At this stage, the variation in results implies difficulty in estimating foil thicknesses. Reducing the pH does give flatter, brighter copper foils. Further investigation is needed to identify the parameters to be set to improve repeatability and achieve reliable foil thicknesses.

![Figure 8: white light interferometry scan, 5mA 2 hours sulphuric acid. Roughness of 53nm achieved](image)

![Figure 9: comparison of plating results with and without sulphuric acid](image)

| Current applied (mA) | Copper thickness (um) | Copper thickness (um) | Copper thickness (um) | Copper thickness (um) |
|----------------------|-----------------------|-----------------------|-----------------------|-----------------------|
|                      | CuSO₄ without H₂SO₄   | CuSO₄ without H₂SO₄   | CuSO₄ with H₂SO₄      | CuSO₄ with H₂SO₄      |
| Side A               | Side B                | Side A                | Side B                | Side B                |
| 5                    | N/A too many nodules  | N/A too many nodules  | 2.5                   | 2.5                   |
| 10                   | 18-20                 | 6-8                   | 20                    | 6                     |
2.6 Characterisation of Foil Roughness

Roughness of the foils deposited was also characterised as this is an important factor for high power laser targets where there is often a tolerance on surface roughness for the foils. It was seen that the roughness improved (Ra reduced) with continuous mixing and with the addition of sulphuric acid. The foils deposited at 5mA current were bright, with a near mirror finish. The foils were characterised using a white light interferometer.

| Current Applied mA | Roughness (Ra)(nm) | Roughness (Ra)(nm) | Roughness (Ra)(nm) | Roughness (Ra)(nm) | Roughness (Ra)(nm) |
|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|
|                    | CuSO4 No H2SO4     | No mixing Side A   | CuSO4 No H2SO4    | Mixed Side B       | CuSO4 No H2SO4    |
|                    | N/A too many nodules| 210                | 180                | N/A delamination   | 53                 |
| 5                  |                    |                    |                    |                    |                    |
| 10                 | 3800               | 1800               | 1000               | 1240               | 240                |
| 15                 | N/A sample too dark to characterise. | 790 | 760 | 400 | 470 |

Table 3: Roughness (Ra) scans using white light interferometry

2.7 Processing Conditions for Optimal Laser Targets

There are many factors that can affect the quality, thickness and usability of plated metals. The five factors investigated here in this preliminary research have shown some improvement in foil success. It can be seen that if layered copper on aluminium targets are required, the best results would be achieved with foils that are electroplated with continuous mixing of the solution and the addition of sulphuric acid, with a seed layer (180nm) and at 5mA. This slow plating rate gave good uniformity and improved surface roughness allowing for adhesive-free, layered metal targets.

From this preliminary investigation there are clear opportunities to improve the quality of the plated foils. Work is planned to investigate the impact of the seed layer thickness. This is important as seed layers also give the opportunity to plate onto non-conductive materials, thus further opening up the option of adhesive-free layered targets. Polishing the aluminium substrate to remove the furrows present in the foils is also an area of further investigation to consider. We hope to develop capabilities to plate metals on to machined structures, which could further reduce the components and assembly in target manufacture.

3. Ultrathin polymer film

We investigated films that are 90nm or less, that are used as a high-power laser target suspended (in most cases) over a hole to produce a membrane target and in this report ultrathin polymer films are made from polyvinyl formal resin. The resin is formed from raw material purchased in solid powder form that is mixed with a solvent to allow coating to take place using dip coating and spin coating methods. Solvent selection included establishing appropriate drying times to eliminate the solvent used in the manufacture process whilst still producing a useful film.

The polymer used for these films has been commercially known as Formvar, (a poly(vinyl) formal) and has traditionally used with TEM grids in microscopy. The original driving force of this work was to produce support films for indirect drive inertial confinement fusion experiments. The support films would sit within the gold hohlraum and support the spherical fuel shell. To satisfy the requirements of
this target the film needs to be thin and strong enough to hold the shell in place without imprinting upon the target when compressed [8]. Thin films have also been used in high power laser experiments looking at efficient proton acceleration via a hybrid scheme of radiation pressure-sheath acceleration in an ultrathin foil irradiated by a linearly polarised laser pulse [9]. Metrics to assess film suitability included looking at film thickness, uniformity across an area, removal from the substrate, and ability to be used as a supporting film.

3.1 Assessing the roughness of the substrate

A profilometer (KLA Tencor Alpha-step IQ) was used to analyse the surface roughness of glass slides. In some cases where the characterisation was not clear due to the resolution limitations of the system, atomic force microscopy was used to assess the roughness. Glass slides that were the smoothest were picked as the substrates in the film study.

![Roughness scan of a microscope slide used in the investigation (Ra = 1.646 nanometers). Scanned on profilometer (KLA Tencor Alpha-step IQ).](image)

3.2 Coating Processes

Preliminary work was carried out to establish a procedure to obtain the thinnest films and both spin coating and dip coating techniques were investigated. Spin coating is a well-established coating technique that employs centrifugal forces to spread a liquid over a spinning substrate. Using spin coating it was found that to produce the best films applying the mixture to the substrate after ramp up (acceleration of the substrate) was ideal. If solution was deposited before the substrate was set to spin the resultant films would exhibit a waviness across the surface. Spin coating did produce thin films; however, they were all above the 90nm and it was not possible to achieve a thinner coating that was uniform across the surface.

Dip coating was the preferred method of film formation because it produced the thinnest films in the preliminary investigation. Films are produced by vertically dipping a flat substrate, in our case glass microscope slides, into the solution and lifting them out at a set rate. The dip rate greatly affects the film thickness, as does the solution concentration. The faster the substrate is pulled out of solution the thicker the resultant film. Once removed from the solvent-resin mixture the slide is left to dry. The solvent quickly evaporates from the slide, leaving behind a film of Formvar. Using this technique, films as thin as 10nm were achievable.

The greatest difficulty encountered during this investigation was removing the film from the glass substrate because as thinner films were achieved they became increasingly difficult to remove from the substrates (this was also apparent with other substrates such as polished silicon wafers). Typical float-off techniques were used, without a release layer, to remove films from the substrates and to help improve ultrathin film release substrates were cleaned using oxygen plasma cleaning. The films were then lifted from the water onto a support frame.
3.3 Characterisation
To characterise the films from both spin and dip coating a surface profilometer is used. Where films are too thin for to resolve the thickness using a profilometer atomic force microscopy is used. The films on the substrate are scored using a scalpel blade to give a step height that can be measured. In other cases, witness slides are masked to give a step for measurement.

Following initial thickness characterisation a second check of the film thickness is made as the films are floated off from the glass substrate onto water and then back on to a clean flat substrate. (This can be another glass microscope slide or polished silicon wafer.) The benefit of this second characterisation step is the substrate will not be damaged from the scoring process giving increased confidence in film thickness characterisation. Film uniformity was also looked at as a factor of success and this was characterized by looking at the thickness across a number of scans as shown in figure 11.

![Figure 11. Lineouts across the edge of a formvar thin film using AFM](image)

Ultrathin Formvar films were successful using dip coating procedures and the thinnest films achievable were 10nm thick. Further work is to be carried out to assess the strength of these films. In addition, work is required to improve the quality of the target frames as roughness of the edges on the mounts used in holding thin films could rupture the films.
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