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ABSTRACT

An improved process for production of ICF Target Mandrels has been developed. Shells made from PAMS (poly-α-methylstyrene) are coated with GDP (glow discharge polymer). The PAMS is then removed by depolymerization and volatilization at 300°C, leaving a GDP mandrel. Compared to past polymer mandrels, this process yields GDP mandrels with significant improvements in wall thickness control, sphericity and concentricity, and the complete absence of vacuoles. The process is capable of making GDP shells with a wide size range (from 300 < o.d. < 2700 μm), and an independently controlled wall thickness (from 1 to 30 μm). The GDP can be doped with a variety of elements.

I. INTRODUCTION

Future ICF experiments require targets with more exacting specifications: larger diameters, thinner walls, improved surface smoothness, wall compositional uniformity, uniform wall thickness, sphericity, etc. No past production method could produce targets with all these parameters being excellent. Recently, Letts *et al*. began work on a depolymerizable mandrel (PAMS) which could be overcoated with GDP, and then the PAMS removed.[1] However, their initial work relied on forming beads of PAMS, which caused subsequent processing problems. They then began using hollow PAMS shells with an o.d. of about 700 μm, and observed improved results. At this point, the work was continued by General Atomics to develop a production process for making target quality shells for ICF experiments.

II. PRODUCTION OF PAMS SHELLS

To produce the starting PAMS shells, we use a droplet generator modeled after the one used at Osaka University's Institute of Laser Energetics.[2] The droplet generator is a dual orifice device, with a water/PVA (polyvinyl alcohol) stripping fluid sweeping the droplets off the orifice and down a collection tube to a stirred beaker. The polymer solution is 11 wt% PAMS (Scientific Polymer Products, 400,000 molecular weight, polydispersity 1.04) dissolved in fluorobenzene (Aldrich). Fluorobenzene is used as a solvent, because it has a density at room temperature of 1.024 g/cc, very similar to the density of the interior water and stripping fluid. With fluorobenzene as a single solvent, the overall density of the polymer solution phase does not change appreciably during curing, avoiding density mismatches which occur during curing with binary solvent solutions. The PVA (Polysciences, 88% hydrolyzed, 25 K mol. wt.) solution is 0.1 wt% in water. Control of the final shell size is dominated by the stripping flow (50–300 cc/minute), and by the collection tube i.d. (4 mm). Shell size is also influenced by the needle sizes. To produce the nominal sized (1000 μm o.d./20 μm wall) dry shells required for the University of Rochester's OMEGA laser, a stripping flow near 300 cc/minute is required. These shells are made at about 30/s, for a period of about 20 minutes. The shells are then cured by heating in a water bath at 60°C for 3–4 h to remove the fluorobenzene solvent. Unless otherwise stated, all discussions below will pertain to shells 900–1000 μm o.d. in size.

Following the curing of the shell wall, the PVA is washed off the shells, leaving a water filled PAMS shell. The water is extracted by placing the shells in a 5–25 vol% ethanol/water solution for 24–48 h. The solution osmotically extracts interior water from the shell. If the osmotic extraction is allowed to continue further, either by using longer times or higher percent ethanol, some 95% of the PAMS shells collapsed, because the smaller volume of

interior water pulls in the shell wall. The osmotic pressure used for extraction is sufficient to buckle the shell wall. However, we found that setting the beaker containing partially extracted shells into a lab ultrasonic bath for 5–60 s was sufficient to nucleate a bubble in most of the thousands of shells and relieve the growing pressure on the shell wall. Upon further water removal, the bubble grows and the pressure drop across the shell wall remains less than one atmosphere, rather than collapsing the shell. Use of the ultrasonic bath improved our shell yield from 5% to about 95%. Final extraction of all the remaining water from the shell is done by heating the shells overnight at 60°C in a vacuum oven. Target quality shells could then be selected for GDP coating.

III. GDP COATING AND PYROLYSIS

Target quality PAMS shells are coated with GDP in the standard plasma polymerization deposition chamber.[3] The GDP coating is deposited according to the particular type of final target needed: wall thickness of each of several layers, each layer being of a different composition.

The GDP overcoated PAMS shells are pyrolyzed under flowing nitrogen in a quartz tube furnace. The PAMS depolymerizes at 300°C to form α-methylstyrene monomer, which then diffuses through the GDP wall and is removed. The pyrolysis is performed according to a protocol developed at Lawrence Livermore National Laboratory (LLNL)[4] as follows:

room temperature to 200°C at 10°C/min
200°C to 300°C at 0.2°C/min
Hold at 300 C for 1000 min

The entire pyrolysis procedure takes one and half days, including furnace cool down time. Hold times as short as 100 min have proven sufficient for fully pyrolyzing and volatilizing the PAMS through 20 μm of GDP overcoating. The fully pyrolyzed mandrels are the final targets made of pure GDP.

The final target may contain multiple layers of doped and undoped GDP. The target may simply contain a single layer of undoped GDP of the desired thickness. Alternatively, the coating may be made up of several layers of varying thicknesses of doped GDP containing any one of the dopants that can currently be incorporated into the plasma glow discharge. Presently, dopants for GDP include germanium (from tetramethylgermane), silicon (from tetramethylsilane), chlorine (from 2-chloro-2-butene), and titanium (from $TiCl_4$). In addition, fully deuterated GDP mandrels can be made using CD_4.

The PAMS technique allows the innermost layer to be a very thin doped GDP layer. This thin layer is not possible at the present time using other methods of target production. For multi-layer shells where the innermost layer is thick (5 or more microns), pyrolysis can be performed after the deposition of any layer. However, when the innermost layer of a multi-layered target must be very thin (1–4 μm), a GDP multi-layer coating of greater than 5 μm must be deposited prior to pyrolysis. This deposition of a thin layer followed by further layers before pyrolysis avoids handling and possibly damaging very thin (< 5 μm) and fragile GDP shells.

The pyrolysis procedure changes the characteristics of the final mandrel in a number of ways. The shell diameter, in general, decreases during pyrolysis. The shrinkage of the shell diameter was also reported in the original work with PAMS beads for undoped GDP.[1] In addition, we have observed that the thickness of the GDP layers deposited also decreases during pyrolysis. We have also found that doped GDP coatings shrink in diameter and wall thickness by different amounts depending on which dopant is present in the GDP. The amount of shrinkage for each type of doped GDP is summarized in Table I. Shells overcoated with silicon-doped GDP do not shrink, apparently due to increased crosslinking. Germanium-doped films, which should be structurally very similar to silicon-doped GDP, do shrink, but less than undoped GDP. For each type of coating, it is critical that the precise amount of diameter shrinkage during pyrolysis is known in order to select initial PAMS mandrels that will result in final GDP mandrels of the desired diameter. The wall thickness shrinkage also neccesitates depositing a slightly thicker GDP layer than is required in the final mandrel. Shorter pyrolysis times at 300°C result in smaller amounts of shrinkage in both diameter and wall thickness. Additional work needs to be performed to find the best pyrolysis protocol to minimize shrinkage while fully pyrolyzing the inner PAMS mandrel.

Most dopants currently used are thermally stable during the pyrolysis, as the dopant concentration is not significantly changed during pyrolysis. Chlorine-doped GDP, however, is thermally unstable. Pyrolysis decreases the chlorine concentration within a layer and chlorine will "bleed" into adjacent layers that had no chlorine prior to pyrolysis. Presumably, HCl is removed from the polymer leaving a double bond, and the shells darken due to conjugation.

The PAMS mandrels are in general spherical on the outside, but non-concentric– there is a wall thickness variation. For 950 μm diameter shells, the non-

Table I
Amount of shrinkage in diameter and wall thickness during the standard pyrolysis of GDP coated PAMS mandrels depends on the type of dopant present in the GDP overcoating

Dopant	Dopant atomic percent	Diameter Shrinkage, %	Wall thickness shrinkage, %
None	- (57% H)	5–6	10
Deuterium	100% D for H (62% D)	5–6	10
Chlorine	various	5–6	10
Ge	2.5	2.2	4
Si	5.5	<1(cannot measure)	< 1(cannot measure)

concentricity is near 10% – the 20 μm wall thickness ranges from 18–22 μm. When these non-concentric shells are bounced gently during GDP deposition, the heavy side is generally on the bottom and a non-uniform GDP coating is produced, and therefore an unacceptable non-concentricity in the final GDP mandrels is produced. In addition, there is a larger spread in the shell to shell wall thickness variation. We can avoid these problems by more vigorous bouncing of the non-concentric PAMS shells during GDP deposition. However, the vigorous bouncing limits the production rate to a small number of shells (~50) in any given run.

IV. SHELL CHARACTERIZATION

The droplet generator makes PAMS shells which are high target quality. For the thousands of OMEGA shells produced, the standard deviation of the o.d. (~960 μm o.d.) is typically 10–20 μm. The average wall thickness is typically 20 μm, and the non-concentricity is 10% or less, demonstrating we have fairly uniform walls (18–22 μm around the shell). The uniform wall thickness permits uniform bounce coating during the GDP coating, to give a more uniform GDP coating. AFM spheremapper traces also show out-of-roundness of less than 1 μm, which corresponds to a mode 1 or mode 2 intensity of 10^4-10^5 nm^2 (Figs. 1 and 2). (An intensity of 10^6 nm^2 would correspond to an amplitude of 1 μm out of round.) These values for 960 μm o.d. shells are excellent, and can probably be further improved upon by lower shear stirring of the shells in the curing bath. Typically, the RMS roughness of the PAMS outer surface is 80–90 nm for modes 2–20, and about 1 nm for modes 20–100. These numbers are "state-of-the-art" values for 900–1000 o.d. micron shells.

The characteristics of the GDP mandrels are similar to those of the typical PAMS shells. The spheremapper AFM

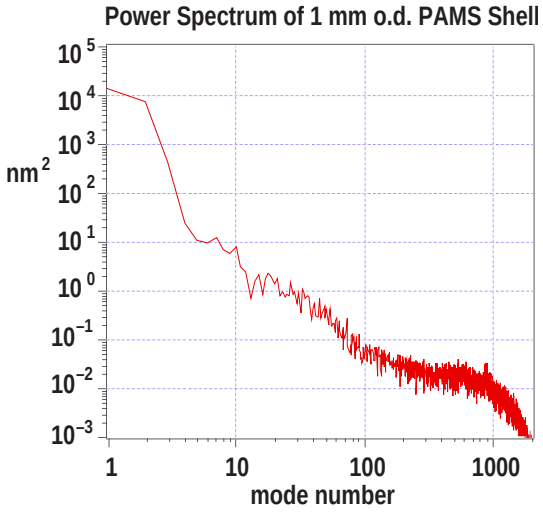

Fig. 1. Power Spectrum of OMEGA-sized PAMS shell.

intensities at mode 1 or 2 are typically 10^4-10^5 nm^2, so the targets are spherical to within less than a micron out of 1000 μm o.d. (Figs. 3 and 4). The RMS surface roughness is about 80–160 nm at modes 2–20, and about 1 nm for modes 20–100. Since GDP coating tends to magnify local surface asperities on the PAMS shell, such low GDP surface roughness is excellent.

V. CONCLUSIONS

The PAMS/GDP process has been demonstrated to form excellent OMEGA shells, in the neighborhood of 900–1000 μm o.d. The process allows greater flexibility in doping. This process is now the basis for all current large target shell deliveries. Several processing complications (have been overcome), such as ultrasonic nucleation of a bubble to prevent osmotic collapse. The degree of shrinkage of GDP mandrels during pyrolysis have been characterized and the pyrolysis process is standardized.

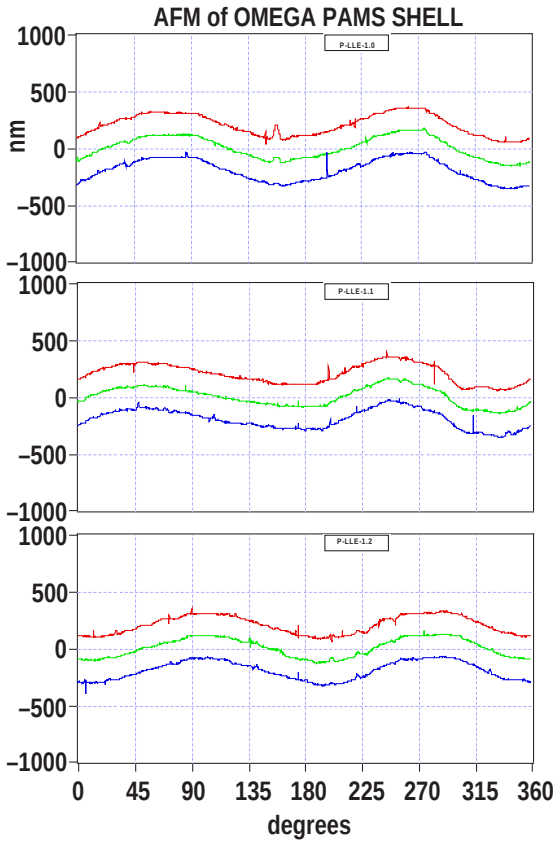

Fig. 2. AFM traces of OMEGA-sized PAMS shell. The traces show the variation in shell radius, as a function of location around the shell. The middle of the three traces is made around the equator, and the displaced traces are scans made above and below the equator. The shell is then rotated 90 degrees, and the same three scans are made. The shell is then rotated in the third orthogonal plane for the third set of scans. All 9 scans are used in calculating the power spectrum of Fig. 1.

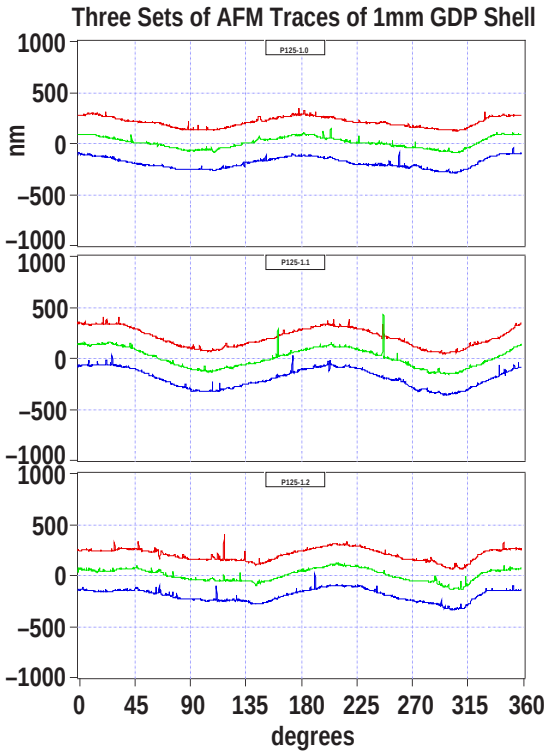

Fig. 3. Three orthogonal sets of three AFM traces of OMEGA-sized GDP shell. Spikes are bits of dust or debris on shells.

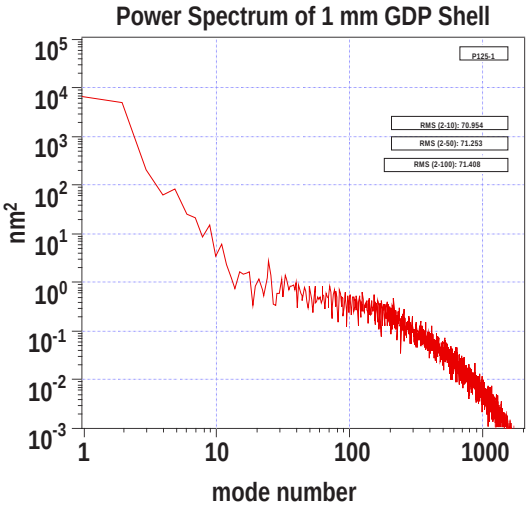

Fig. 4. Power Spectrum of OMEGA-sized GDP shell.

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