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Production of Higher Strength Thin Walled Glow Discharge Polymer Shells for Cryogenic Experiments at OMEGA

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Thin walled polymer shells are needed for OMEGA cryogenic laser experiments. These capsules need to be about 900 μm in diameter and as thin as possible (≈ 1–2 μm), while having enough strength to be filled with DT as fast as possible to about 1000 atm. We have found that by optimizing the coating parameters in the glow discharge polymer (GDP) deposition system, traditionally used for making ICF targets, we can routinely make robust, ≈ 1.5 μm thick, 900 μm diameter GDP shells with buckle strengths of over 0.3 atm. This is twice the strength of shells made prior to the optimization and is comparable to values quoted for polyimide shells. In addition, these shells were found to be approximately three times more permeable and over 20% denser than previously made GDP shells. The combination of higher strength and permeability is ideal for direct drive cryogenic targets at OMEGA. Shells as thin as 0.5 μm have been made. In this paper, we discuss the shell fabrication process, effects of modifying various GDP deposition parameters on shell properties and chemical composition.

1. Introduction

Polymer capsules, about 900 μm in diameter and as thin as possible (≈1 μm), are needed for direct drive OMEGA cryogenic laser experiments. These shells have to be filled with D_2 or DT as fast as possible to about 1000 atm. The maximum fill rate is given by P_b/τ, where P_b is the shell buckle strength and τ is the gas permeation time constant. Therefore, for the fastest filling rates which ensures low tritium inventory and ^{3}He buildup, high aspect ratio shells with high buckle strength and permeability are required. In addition, these shells have to be strong enough to survive routine handling required for characterization and assembly. Fabrication of thin wall capsules in the OMEGA size range has been demonstrated by other groups using vapor phase deposition of polyimide [1] and emulsion [2] techniques. We have used the depolymerizable mandrel technique [3] for fabrication of such shells. Our shells have recently been used in some of the first cryogenic shots at OMEGA [4]. In this paper, we review our previous work [5,6] and present new results particularly fabrication of fully deuterated thin wall shells.

2. Depolymerizable Mandrel Technique

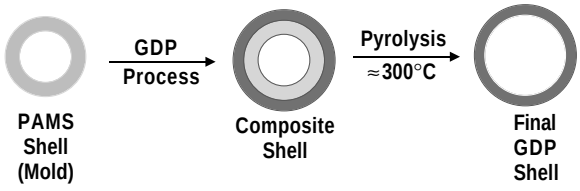

Fig. 1. GDP shells are made using the depolymerizable mandrel technique.

The depolymerizable mandrel technique is currently used extensively in fabrication of most ICF capsule targets in the U.S. [7]. The process is illustrated in Fig. 1. High quality Poly-alpha-methylstyrene mandrels are selected to have excellent sphericity (99.9%) and wall non-uniformity (<5%). These mandrels are over-coated with a GDP coating of the desired thickness. The deposited GDP coating is very uniform as well, as the shells are continually agitated during deposition. Upon pyrolysis at 300°C, the PAMS mandrel depolymerizes into its monomer vapor, which diffuses out of

the GDP coating, which is stable at that temperature. This leaves a final capsule which is made up of the GDP coating alone. We have extended the depolymerizable mandrel technique process to fabricate thin wall shells by limiting the coating in thickness to a few microns.

3. Optimization of GDP Coating Parameters

3.1. Buckle Strength

The material properties of the final GDP capsule, including those crucial for the OMEGA target filling process, namely buckle strength and permeability, strongly depend on the GDP coating conditions. Therefore, we investigated the effects of varying the GDP deposition conditions on the properties of the final GDP shells. In these studies, we found that shell strength depended strongly on two particular parameters: deposition pressure and process gas flow ratios. Shells with highest buckle strengths were obtained at the lowest deposition pressures. Reduction of coating pressure from the normal setting of 75 mTorr produced stronger shells. However, below a minimum deposition pressure of about (25 mTorr) the PAMS mandrels were etched, sometimes so severely that they disintegrate during deposition.

The process gases normally used for plasma polymer deposition for ICF target fabrication are hydrogen and a hydrocarbon gas such as trans-2-butene (T2B). Hydrogen acts as an etching agent during deposition, while plasma disintegration of T2B in the glow discharge produces fragments that form the polymer. Although, T2B is used extensively in fabrication of thin wall shells, other hydrocarbon precursors, such as methane and benzene, have also been used. The flow ratio of hydrogen and the hydrocarbon precursor is the other critical parameters in obtaining strong thin wall shells. Figure 2 shows the dependence of buckle strength of shells on the deposition pressure and the relative flow rates of hydrogen and methane. As the hydrogen flow is increased the resulting shells become stronger. We beleive that etching by hydrogen at lower deposition pressures removes weak links and leads to a more strongly bonded matrix, as is the case in diamond-like coatings [9]. The calculated Young's Modulus is used as a measure of buckle strength of shells to properly compare shells made using different conditions, which may have aspect ratio differences. This is done because the theoretical buckle strength of an ideal defect-free spherical shell of wall thickness w and diameter d, given by the expression [10],

$$E = \frac{\sqrt{3\left(\nu^2 - 1\right)}}{8} P_b \left(\frac{D}{w}\right)^2 \quad , \tag{1}$$

depends strongly on shell aspect ratio, D/w. Here, E and ν are the Young's Modulus and Poisson's ratio for the material, respectively. Therefore, calculation of Young's Modulus properly accounts for dimensional differences between various shells and can be used for comparison of buckle strength of shells.

By optimizing the above GDP coating parameters we can now routinely make thin wall shells which are more than a factor of two stronger than "normal" GDP shells, i.e. those produced prior to the optimization. The buckle strength of these stronger shells is 1.5 atm for 3 μm thick, 900 μm diameter shells. Figure 3 shows an example of a batch of shells which have a wall thickness of about 1.3 μm.

The optimization of the coating parameters to obtain stronger shells did not have a deleterious effect on the surface finish of the shells. In fact, the surface finish improved as the

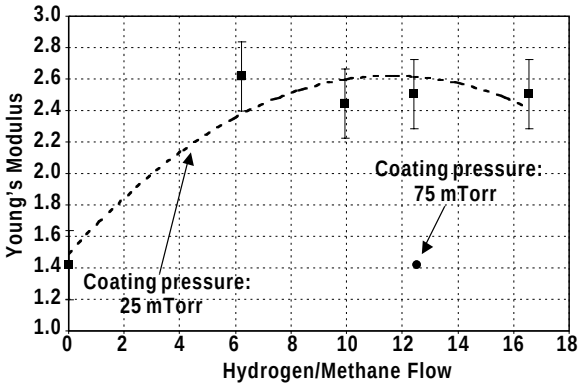

Fig. 2. Modification of coating pressure and gas flow ratios had the largest effect on shell buckle strength. Lower deposition pressures at higher hydrogen to hydrocarbon gas (methane in this case) led to stronger shells (as measured by the Young's Modulus).

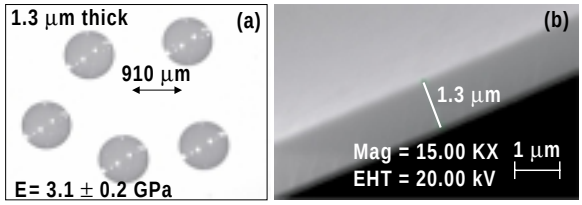

Fig. 3. (a) Robust thin wall GDP shells (1–3 μm) can be made routinely using the depolymerizable mandrel technique. (b) Scanning electron microscope image of the wall of a 1.3 μm thick shell.

hydrocarbon flow was decreased with respect to H_2 flow. The surface finish improvement with decreasing T2B/ H_2 flow ratio, well known in our GDP deposition system at higher deposition pressures, was therefore also effective at the lower deposition pressures as well.

2.3. Permeability

Permeability measurements on numerous shells made using T2B against various gases such He, Ne, N_2 and Xe indicate that the stronger shells made using optimized coating conditions are in fact several times more permeable than "normal" GDP shells. Independent permeability measurements of these shells at UR/LLE confirmed the higher permeability of the stronger shells. The combination of higher permeability and buckle strength is ideal for the OMEGA cryogenic campaign. As mentioned previously, these shells have indeed been fielded in the OMEGA cryogenic system and imploded. The D_2 permeability still needs to be determined for these shells. However, due to the very short time constant (<9 s for a 3 μm thick, 900 μm diameter shell) of this gas, the measurements will be difficult.

3.3 Chemical Composition

We determined the chemical composition of our films using combustion analysis. The carbon-to-hydrogen ratio in the stronger shells is about 1.16 while for normal GDP (lower strength) it is only 0.85. The lower hydrogen content may explain the higher strength of shells made using optimized coating conditions. The oxygen content of the higher modulus films was also higher (5.5 at. %) compared to normal GDP (3.0 at. %). The density of fresh (minimal exposure to oxygen) higher modulus films was measured to be 1.20 g/cc compared to 1.04 g/cc for normal GDP. This is again consistent with the higher carbon content of higher modulus films. This higher density can be of value for indirect drive NIF capsules as well. Film composition was also examined by infrared spectroscopy and the lower absorption in the CH band confirmed the lower hydrogen content of the stronger shells (Fig. 4) [11].

3.4. Deuterated Coatings

Infrared (IR) radiation is used to layer D_2 ice layers inside polymer shells. It is also being studied for enhanced layering of DT ice as well. Fully deuterated shells may be needed in order to avoid significant absorption of the IR radiation in the capsule wall as opposed to the fuel ice layer. We have therefore extended our process to fabricate deuterated shells. The hydrogenated process gases, hydrogen and T2B, were replaced with their deuterated analogs,

deuterium and deuterated T2B. It was found that optimized flow setting for deuterated coating was quite different than those found for the CH coatings. The optimization procedure had to be repeated for the deuterated gases. Thin walled deuterated shells made using these optimized conditions also had twice the buckle strength of those made prior to the optimization. These deuterated shells are also currently being used for cryogenic shots at OMEGA.

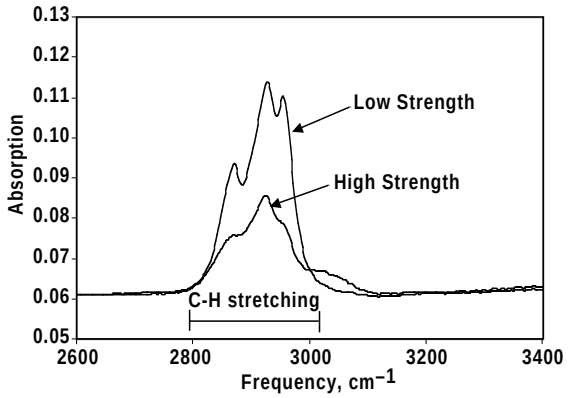

Fig. 4.. Robust shells made by optimization of GDP coating parameters contain less hydrogen as seen in the infrared absorption spectrum shown in the figure. The lower hydrogen content implies a higher degree of cross-linking which explains the higher strength of these shells.

4. Conclusion

We have made major advances in developing strong thin-walled GDP capsules. By optimizing the coating parameters, we can achieve over a factor of 2 increase in the normalized buckling pressure (0.3 atm for a 1.5 μm thick, 900 μm diameter shell) compared to the usual GDP capsules. This high strength material is several times more permeable than normal GDP to a number of different gases. Shells made from this material can therefore be permeation filled several times faster. The strength of this high-strength GDP is comparable to that of vapor deposited polyimide. The good surface finish that can be achieved with GDP capsules is retained with the high strength material. These higher strength coatings are denser than "normal" GDP and may be useful for indirect drive capsule material, where higher density may lead to better performance.

Acknowledgments

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