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FABRICATION OF THIN WALLED GLOW DISCHARGE POLYMER SHELLS

A. Nikroo and J.M. Pontelandolfo
Inertial Fusion Division, General Atomics
P.O. Box 85608
San Diego, Ca 92186-9784
(619) 455-2931

ABSTRACT

Thin walled (≈ 1 μm) plastic shells, about 900 μm in diameter, are needed for the OMEGA cryogenic experiments. We investigated the possibility of fabricating these targets by modifying the coating parameters in the glow discharge polymerization system traditionally used for making ICF targets. The final plasma polymer shells were tested for buckling pressure in a home-made apparatus. Robust 1 μm thick shells with buckling pressures above 0.1 atm (1.5 psi) could be routinely made by depositing at lower system pressures. Effects of some other deposition parameters are also discussed.

I. INTRODUCTION

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 μm), while having enough strength to be filled with DT as fast as possible to about 1000 atm. This places stringent strength requirements on the material the shell is made from. If the shells are filled at a constant rate, then they will survive if the fill rate is less than P_b/τ, with P_b being the buckling pressure of the shell and τ the permeation time constant. Therefore, shells with greater buckling pressures can be filled faster. In addition, these shells must be robust enough for rather routine handling. Recently, vapor phase deposition[1] and emulsion [2] techniques have been used for fabrication of thin walled shells. In this study, we investigated the possibility of making thin walled shells using the glow discharge polymer (GDP) process, by exploring the coating parameter space of the helical resonator system[3] used for depositing such films. This study was not meant to comprehensive but rather preliminary with the hope of continuing the work in the future.

II. EXPERIMENTAL

Thin walled GDP shells were made by the depolymerizable mandrel technique using poly-alpha-methylstyrene (PAMS) mandrels.[4] The GDP deposition system hardware has been described previously in the literature.[3] It is used on a routine basis to fabricate capsules for ICF experiments.[5] In this process, trans-2-butene is mixed with hydrogen, the mixture is broken down in the plasma region and the hydrocarbon fragments deposit on the substrates forming ultrasmooth coatings. The flow rates and plasma parameters usually employed in fabricating ICF targets are listed in Table I. Table I also shows the range over which the deposition parameters were varied in this study. These parameters were not all independent. For example, as the H_2 flow was increased, the minimum system pressure also increased. Therefore, it was not possible to perform a coating run at high H_2 flows and low system pressures. In addition, the PAMS shells disintegrated during the coatings at low pressures and high powers. Therefore, this also limited the usable parameter space. In a number of cases multiple runs using the same coating conditions were performed to verify the reproducibility of the coatings.

Making a GDP shell using the depolymerizable mandrel technique involves a number of steps and a number parameters that can be varied in each step. We decided to vary only the GDP depositions parameters in our initial study reported here, to reduce the parameter space as much as possible. Therefore, all thin walled shells tested were made by depositing GDP on PAMS mandrels from the same batch. After deposition they were stored under vacuum until pyrolysis to avoid oxygen uptake in the film.[6] They were pyrolyzed using the same furnace and the same pyrolysis conditions. They were all tested shortly after the pyrolysis step to avoid prolonged exposure to air.

Table I
Summary of the coating parameters which were varied and the range over which they were varied. Trans-2-butene flow was kept constant during experiments

Parameter	Usual Value	Range Examined
Coating Pressure	75 mTorr	25–200
RF Power	12.5 W	3-30 W
Hydrogen Flow	10 sccm	2–10 sccm
Trans-2-butene	0.15 sccm	—

It was crucial to characterize the shells accurately and obtain enough statistics to make valid comparisons between shells made using different coating conditions. This involved determining shell aspect ratios and buckling pressures. Wall thickness measurements were made by interferometry and occasionally verified by SEM measurements. Shell wall thickness could be measured to better than ≈ 10%. Diameter measurements had an error of about 0.5%. Only shells from batches with wall thickness uniformities of better than 10% were tested. An apparatus to measure the buckling pressure of our shells was made as shown in Fig. 1. Shells were placed in a grid inside the main chamber, which had windows for direct optical inspection of shells. This main chamber was connected to a gas supply and a vacuum pump. Buckle testing was performed by overpressurizing the chamber suddenly (< 1 s) with nitrogen or SF_6 which have very low permeabilities through GDP (half-lives > 3 min for 900 μm × 1 μm shells). After quick inspection of the shells for survival or failure after pressurization, the chamber was vented rapidly to avoid filling shells with the test gas. This procedure was repeated with increasing the overpressure in small increments until the shells buckled. A pressure regulator with 0.5 atm (7 psi) range and 0.007 atm (0.1 psi) resolution was used for most of the measurements.

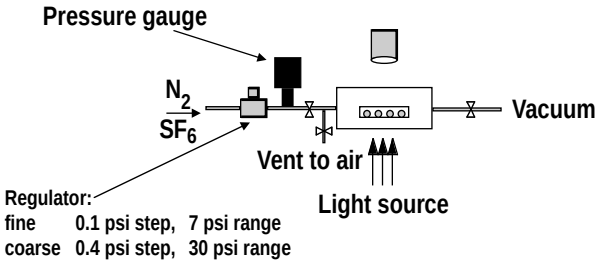

Fig. 1. Schematic of the buckle testing system. Nitrogen or SF6 were used as the test gases due to their low permeability through GDP. For many of the tests a fine control pressure regulator was used with a step resolution of 0.1 psi. In tests involving pressures higher then 7 psi, a coarser regulator was used. Shells were subjected to the overpressure suddenly (<1 s) and the overpressure was removed quickly to avoid filling the shells with gas. The onset of buckling was determined by observing the shells via the optical access.

III. RESULTS AND DISCUSSION

Theoretically, for defect free shells, the buckling pressure, P_b, is given by:[7]

$$P_b = K \cdot E \cdot (t/d)^2 \quad (1)$$

where E is the Young's modulus, d is the shell diameter, and t is the shell wall thickness. K is a constant. Ideally, for accurate comparisons of buckling pressures, it is desirable to make shells of the same exact thickness and diameter in all the various coating runs. However, in practice it is almost impossible to do that in a timely fashion. Therefore, to perform a comparison of buckling pressure of shells made under different conditions, any difference in the aspect ratio, (d/t), needs to be properly accounted for. To do this, we normalized all measured buckling pressures of shells of varying aspect ratios to that of a 900 μm diameter, 1 μm thick shell. To test the validity of this procedure we made shells of three different wall thicknesses in the same extended coating run by removing shells at two different times during the coating. The results are shown in Fig. 2. As can be seen in the figure, even though a slight deviation is observed, the scaling expected in Eq. (1) appears to be valid within the measurement uncertainties.

Changing the system pressure had the most profound effect on the strength of the shells. The pumping speed available in our system limited the lowest possible pressure obtainable in our system to about 25 mTorr. The chamber pressure was measured close to the location of the shells within the chamber. Coatings at this pressure resulted in the strongest shells as shown in Fig. 3. We

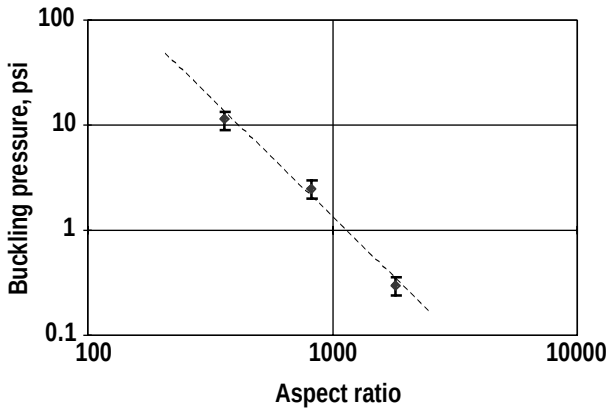

Fig. 2. Plot of the buckling pressures of shells of three different thicknesses made under virtually identical conditions. Buckling pressures of these shells scale inversely with the square their aspect ratios as expected from Eq. (1). This provides validation for normalization procedure used for comparing buckling pressures of shells with different aspect ratios.

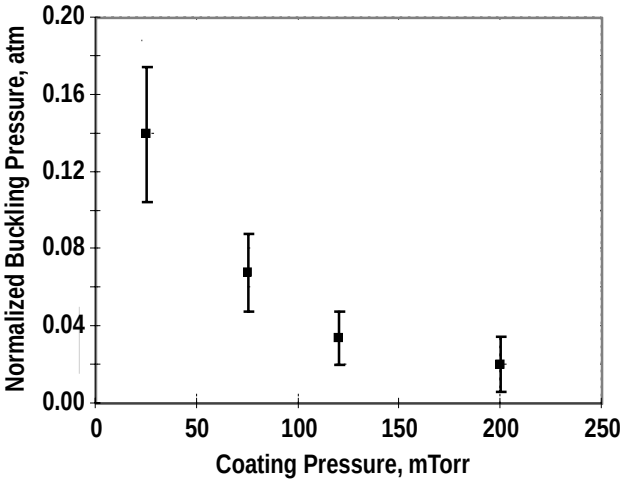

Fig. 3. Lowering the system coating pressure leads to thin walled shells with greater buckling pressures. The rf power, the hydrogen flow, and the trans-2-butene flow were kept constant at 12.5 Ws, 2 sccm, and 0.15 sccm, respectively, in all the runs. Over ten runs were made at 25 and 75 mTorr and the data represents values obtained on over 50 shells. The error bars are a combination of the measurement errors and the variations in the observed buckling pressures of shells.

made numerous runs under these conditions in two separate coating systems, all resulting in shells that had normalized buckling pressures of about 0.140 ± 0.035 atm (2 ± 0.5 psi). In contrast, shells made using deposition parameters usually used for fabricating ICF capsules (75 mTorr) only had a normalized buckling pressure of ≈ 0.07 ± 0.02 atm (1.0 ± 0.3 psi). There was a rather large scatter in the observed buckling pressures of shells from a given batch. Shells from a given batch could have buckling pressures varying by over 25%, presumably due to defects or slight wall non-uniformities. For each batch the average of the observed buckle pressures was taken to represent the value for that batch. The data points in the graph represent the average of values from many batches. For example, the points at 25 and 75 mTorr each represent over 40 shells from about eight different batches. The error bars represent the measurement errors discussed previously.

The stronger shells made using lower deposition pressures (LDP) have a number of features that are very different from shells made using our usual higher deposition pressures (HDP). The LDP shells are much more robust in handling. For example, they are robust enough to be easily spheremapped[8] by the atomic force microscope (AFM) (Fig. 4). The AFM spheremap indicates no major wrinkling of the outside surface despite the very large aspect ratio. The overall surface finish is good, with the higher modes (> 100) being typical of the GDP process, and the lower order modes (< 100) resembling that of the original PAMS mandrels. The LDP shells are also noticeably darker in color and their buckling behavior is also very different from that of the HDP shells. The LDP shells buckle irreversibly, often

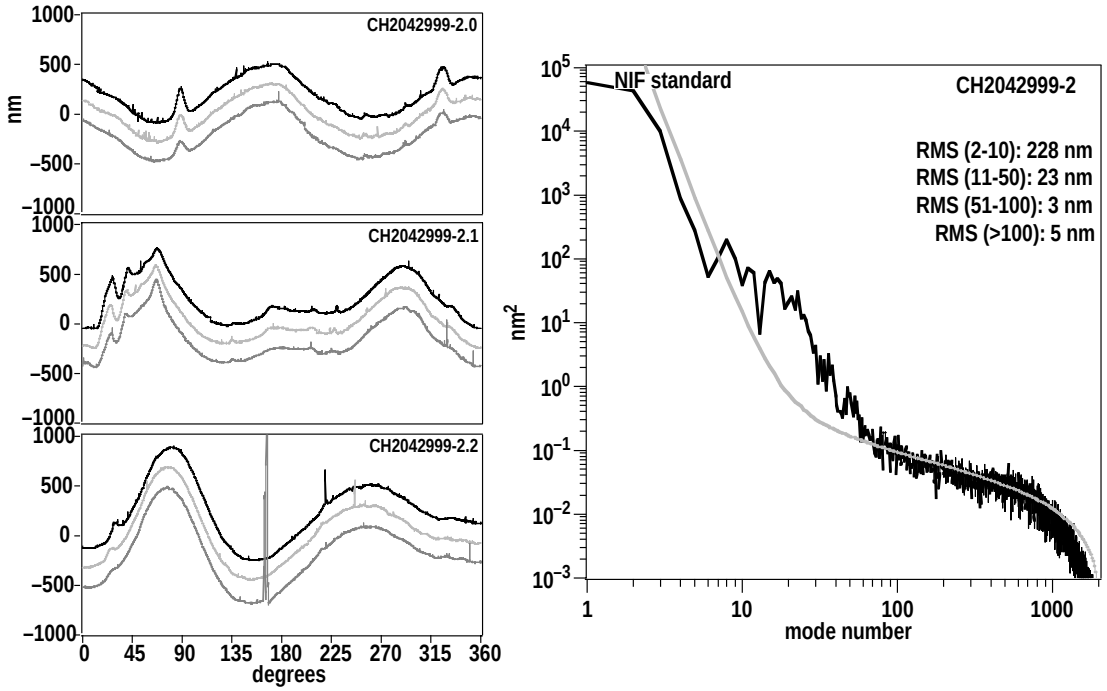

Fig. 4. AFM spheremap of a ≈ 1.1 μm thick, 930 μm diameter shell. The traces on the right reveal the absence of any major wrinkling of the surface that might be expected for a very high aspect ratio shells. The power spectrum on the left resembles that of an original PAMS mandrel used in making the shell.

shattering or tearing when they buckle, as shown in Fig. 5(a). HDP shells are more elastic. Figure 5(b) shows a HDP shell buckled under 2 psi of overpressure. Figure 5(c) shows the same shell, after the overpressure is removed. The HDP shell has bounced back. This underscores the importance of real time optical monitoring of shells during the buckling pressure measurements.

Coating pressure is a key parameter in all types of plasma deposition techniques. It controls the mean free paths of the molecules in the discharge and therefore their kinetic energies. For example, lowering system pressure leads to denser films in sputtering processes as well. This may provide a partial explanation for what we observe as well. However, other effects such as possibly higher plasma densities near the substrates at lower pressure may also contribute to making stronger films.[9] Lowering the pressure in our system led to increased residual stress in the films as well. This could be inferred from the fact that the coating deposited on the plasma tube flaked off after a relatively short LDP deposition time (4 hrs) compared to what is usually observed in HDP runs (over 150 hrs). However, for such thin coatings, the majority of the residual stress in the coating on shells is more than likely relieved during the pyrolysis when the PAMS inner mandrel is removed. We hope to examine the shells by analytical methods other than simple buckle testing to gain some insight into possible structural differences in films deposited at different pressures.

Varying the other parameters over the ranges studied did not result in stronger shells. Varying the hydrogen flow at the same pressure did not appear to have a significant effect on the buckling pressure of the resulting shells. Lowering the radio frequency (rf) power did result in shells which had lower buckling pressures. Increasing the rf power beyond the usual 12.5 W at a pressure of 25 mTorr resulted in the cracking, etching, or complete disintegration of the PAMS shells. The PAMS shells did survive increasing the rf power at higher system pressures, but the resulting shells were not as strong as those made at the lower pressures and 12.5 W.

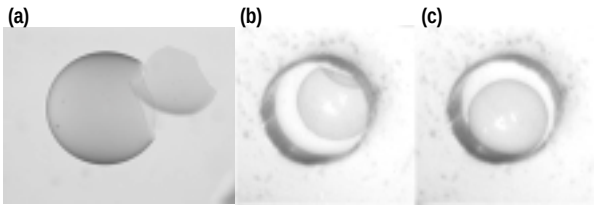

Fig. 5. (a) A LDP shell after having been buckled. The LDP shells are darker in color buckle irreversibly unlike HDP shells. Often they develop a tear when they buckle. (b) A HDP shell inside the buckle testing apparatus under 0.14 atm (2 psi) of overpressure. (c) The same shell after removal of the overpressure. The shell has bounced back. Without optical access the buckling of the shell at this overpressure would not have been noticed.

IV. CONCLUSION

Thin walled polymer shells can be made by GDP deposition using the depolymerizable mandrel technique. A preliminary study of the effects of varying the coating parameters was performed. Lowering the system pressure produced the most dramatic effect. Robust shells which could be routinely handled were made at low system pressures. These have buckle pressures of approximately 0.140 ± 0.035 atm. There could be over 25% variation in the buckling pressure of shells from the same batch and about as much between batches. At the present, we do not have a knowledge of the permeation time constant of these types of shells. This parameter needs to be determined to properly evaluate these shells for use in the cryogenic experiments. We plan to continue this work and explore other parameters involved in our system, in particular, the configuration of the helical resonator plasma generator, which should have a substantial influence on the properties of the GDP coatings.

IV. ACKNOWLEDGMENTS

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