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Low-density foam shells are currently being employed as direct drive targets on the Omega laser facility at the University of Rochester. For cryogenic shots, only a thin layer of glow discharge polymer (GDP) is required over these foam shells to hold the  $D_2$  (or DT) fill provided the capsules are re-filled after cooling. Room temperature surrogate experiments, however, require an additional permeation barrier of aluminum on GDP coated foam shells. This barrier should have a permeation time constant of at least 4 h for  $D_2$  at room temperature. To study this coating, 0.1 um layers of Al were deposited via magnetron sputtering onto the surface of GDP shells and GDP coated foam shells. The foam shells were 180 mg/cc resorcinol formaldehyde (RF) with a GDP thickness of 3-5  $\mu$ m; the GDP shells used for this study had a wall thickness of 25-30 µm. Preliminary data shows that the permeation rate of  $D_2$  for smooth GDP shells is lower than for GDP coated RF shells with a similar thickness of Al. The main factor in this difference appears to be the surface roughness of the shells.

### I. INTRODUCTION

A type of direct drive inertial confinement fusion design consists of a foam capsule with a cryogenic  $D_2$ layer extending inward beyond the foam layer.<sup>1</sup> For near term experiments, however, it is advantageous to have a room temperature surrogate target instead. One of the advantages of the surrogates is that less effort is required to fill and field for the experiment so that more data can be collected in a given amount of time. This surrogate target is a ~900 µm diameter shell consisting of a highdensity (180 mg/cc) resorcinol formaldehyde (HDRF) shell with a 4-5 micron glow discharge polymer (GDP) capping layer. Finally a ~0.1 micron layer of sputtered Al is over coated on the shell to serve as a permeation barrier for D<sub>2</sub> at room temperature. Figure 1 shows a cross section of such a target. The permeation loss of D<sub>2</sub> from this target can be described in terms of an exponential with a given time constant. In order for the capsule to retain the fill gas during mounting and transport to the target chamber, it is necessary for the capsules to have a  $D_2$  permeation time constant greater than 4 h.



Figure 1. Spherical sector of aerogel direct drive capsule for room temperature (cryogenic surrogate) shots. The  $0.1 \mu m$  thick aluminum coating forms the outermost layer of the target.

Much work is invested in GDP coated foam shells prior to Al coating, so it is of particular interest to increase the yield of shells that can hold the D<sub>2</sub> fill. After GDP and Al coating the yield of foam shells that fulfill all the specifications and hold gas with a 4 h time constant is less than 1%. Since the permeation rate of  $D_2$  through the capsule is dominated by the aluminum coating, it is likely that the interface between the aluminum and the plastic coating strongly influences the permeation. This paper investigates the effects that GDP surface roughness has on the permeability of  $D_2$  through a thin (~0.1 µm) magnetron sputtered Al layer. By using GDP shells without the foam, the roughness of the shells could be better controlled allowing a better comparison of roughness and permeation rates. This data can then be compared against that of a GDP coated HDRF shells of varied roughness. Details on the Al coating technique as well as GDP surface characterization will be presented and correlated to the D<sub>2</sub> permeation time constant.

### **II. EXPERIMENTAL**

Full density GDP mandrels of various surface roughness and wall thickness ranging from 25-30 µm were prepared for this study. In addition high-density RF foam shells with a 4-5  $\mu$ m capping layer of GDP were fabricated and investigated. The roughness of the GDP layer was measured using the Veeco NT3300 instrument, an optical profiler for 3D surface topography measurements. All shells used for this study spent the majority of their time under vacuum or filling with D<sub>2</sub> in a high-pressure fill tube. Their exposure to air was kept to a minimum.

Full density GDP shells are fabricated by overcoating PAMS (poly- $\alpha$ -methylstyrene) mandrels with GDP by a plasma enhanced vapor deposition process. During the GDP deposition, the mandrels are agitated by bouncing in a pan to provide a uniform coating over the entire surface. The PAMS is then removed by depolymerization and volatilization at 300°C, leaving a freestanding GDP shell.<sup>2</sup> HDRF aerogel shells are fabricated by a microencapsulation process using a triple orifice droplet generator. The wet shells are then dried with a supercritical CO<sub>2</sub> drying process, and finally a thin GDP capping layer on the order of 4-5 microns is coated over the shell.<sup>3</sup>

Prior to Al coating the shells were placed on Gel Pack, a sticky surface that keeps the shells stationary during coating without leaving a noticeable reside on the shell. A magnetron-sputtering source was used to deposit the Al layer. The background pressure in the chamber prior to sputter deposition was on the order of  $\sim 10^{-6}$  torr and was backfilled with 5 mtorr of argon. A direct current power supply was used to supply 100 Watts to the magnetron-sputtering gun. The substrate to source distance was 12 cm, and the temperature of the substrates reached 85°Ce during coating. Figure 2 shows the typical setup for such a coating.



Figure 2. Typical Al coating setup. Distance was set at 12 cm.

Previous work done on Al permeation barriers studied the effect of coating parameters on  $D_2$  gas retention.<sup>4,5</sup> Stalk mounting of full density GDP shells was used in these test cases. Stalk mounting the shells was not used as an agitation mechanism during the Al coating in this work since GDP coated foam shells are extremely fragile and are prone to cracking under such handling. In addition the yield of Al-coated foam shells with a sufficient permeation time constant is very low, so coating many shells at once on gel pack is a time saving process over stalk mounting and Al coating one shell at a time. A flip coating technique is used to coat Al on the outer surfaces of shells (Fig. 3). Once one side of the coating is completed they are flipped over using a vacuum chuck to expose the uncoated GDP region that was previously in contact with the Gel Pack. An optical microscope is used to align the shells in the proper orientation prior to each coating. Surrogate full density GDP shells act as a witness when coated along side GDP coated foam shells.



Figure 3. High-density RF foam shells and full density GDP shells are magnetron sputter coated with Al using a 2-step flipping technique. Surrogate full density CH shells act as a witness for gas retention measurements.

Following Al deposition the shells were removed from the Gel Pack and inspected for dents, cracks, or any other features that would render the target unusable. Following this step they were placed in a pressure vessel and were filled to 2.0 atm at room temperature using  $D_2$ as the fill gas. The time constant was measured using a mass spectrometer made specifically for measuring spherical time constants of ICF targets.<sup>6</sup>

#### **III. RESULTS AND DISCUSSION**

Since the shell sits stationary during the Al coating, a thickness gradient is established across the surface of the shell. During coating the poles of the shell are coated more thickly than the equator of the shell. To adequately resolve the thickness non-uniformity of the deposited Al layer, a subset of shells were coated beyond the target specification to ~18  $\mu$ m (at the thickness pole region) for the purpose of falling within the measurable bounds of contact radiography.<sup>7</sup> The final thickness was found to deviate by ~50% around the shell (Fig. 4). From this data we expect the coating to vary from 500-1000 Å around the shell of an actual target, with the equatorial region having thinnest Al layer.



Figure 4. (a) Radiograph image used to measure thickness non-uniformities of the sputtered Al layer. (b) Radiography data was taken on a shell with only one side coated Al coated. The data was inversed and the sum of the two sets was found giving the Al layer thickness deviation around the shell in one plane.

Four types of GDP surfaces with differing roughness were used in this study: (a) GDP shells that had a fairly smooth surface finish on the order of 20-30 nm RMS, (b) shells with many isolated dome defects, (c) shells with an increased background roughness, and (d) GDP-coated HDRF shells. The surface roughness on the shells with many isolated defects were produced by vigorous pan bouncing during the GDP coating process in a plasma enhanced chemical vapor deposition (PECVD) system. This created shells with many isolated defects on the order of 1-5 microns in diameter, and an RMS roughness of ~60 nm. In order to fabricate shells with a high frequency roughness, plasma etching was used. For plasma etching an identical apparatus to that used for the GDP coating was used, but without the introduction of a hydrocarbon gas. Plasma etching for ~17 h produced shells with an increased background surface roughness, with an RMS roughness of ~44 nm. Figure 5 shows optical profiler images as well as optical photographs of the various surface finishes.

In general, the smooth plastic shells coated with aluminum had longer permeation rates than similar GDP-coated RF shells, as shown in Fig. 6. The major difference between these two types of shells was surface roughness. Full density GDP shells have a RMS roughness of  $\sim$ 20 nm as compared to GDP-coated foam shells which have an RMS roughness of greater than 50 nm.

To investigate roughness more carefully, a series of full density shells with controlled roughness were measured (Fig. 7). As discussed earlier in Sec. II, three major classes of surface finishes were produced: smooth shells, shells with high frequency roughness, and shells with many isolated defects. The smooth GDP shells coated with Al consistently had a longer permeation constant than similar shells with a rougher surface. It appeared that this was independent of the type of surface roughness. The shells tested with a roughness dominated by large isolated defects had similar D<sub>2</sub> permeation rates as those with higher frequency roughness.



Figure 5. Shells of varying roughness were produced and Al coated to test the roughness effect on  $D_2$  permeation. The images above are contour plots from optical profiling data (with the curvature of the shell removed), and the images in the bottom row are pictures from an optical microscope.



Figure 6. The  $D_2$  permeation rate of shells from 5 separate aluminum coating runs. The full density CH shells (squares) typically had a higher time constant than foam shells (circles), independent of the Al coating batch.



Figure 7.  $D_2$  permeation time constant after Al coating of 3 different sets of GDP shells with different roughness. The smooth shells (A) consistently had a longer permeation time constant than the rough shells (B and C).

Also investigated was how the surface roughness of GDP coated foam shells affected the D<sub>2</sub> time constant. The data in Fig. 8 data shows that foam shells with a smoother GDP surface held D<sub>2</sub> more effectively than rougher shells. Those shells with a GDP RMS roughness of less than 50 nm held gas 80 % of the time above the 4 h D<sub>2</sub> time constant design specification. In comparison those GDP coated foam shells with an RMS roughness of greater than 50 nm had a 0% yield of shells meeting the 4 h time constant specification (Fig. 8). Although the smoother shells had a much better yield of shells with an acceptable permeation rate, there is a wide range of time constants. This suggests that there may be other variables involved such as accuracy of the inversion of the shells during the flip coating method, debris on shells, or local variations in smoothness of the shells.

#### **IV. CONCLUSION**

Gas retention measurements showed a correlation between the variations in GDP surface and  $D_2$  permeation of the Al coated surfaces. From comparisons between



Figure 8. Roughness vs.  $D_2$  time constant for some Al coated HDRF shells. Rougher foam shells had a lower yield that met the 4 h  $D_2$  time constant requirement.

shells with different surface roughness, the trend shows GDP shells with a rougher surface and more isolated defects do not hold gas as well as those GDP shells with a smoother surface. Although other factors that may affect the permeation rate have not been eliminated, the data indicates that starting with smoother surfaces can provide better gas retention in these Al coated shells.

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#### REFERENCES

- S. SKUPSKY, et al., "Advance Direct-Drive Target Designs for NIF," *Third International Conference on Inertial Fusion Sciences and Applications* (IFSA 2003); Am. Nucl. Soc. Inc., La Grange Park, Illinois (2004) p. 61.
- B. W. MCQUILLAN, et al., "The PAMS/GDP Process for Production of ICF Target Mandrels," *Fusion Technol.* 31 (1997).
- R. PAGUIO, et al., "Improving the Wall Uniformity of OMEGA Sized Resorcinol Formaldehyde Foam Shells by Modifying Emulsion Components," *Fusion Sci. Technol.*, this issue.
- M. D. WITTMAN, et al., "Controlling the Permeability of Shinethrough Barriers on Inertial Fusion Targets," Presented at the 12th Annual Target Fabrication Specialist Meeting, Jackson Hole, Wyoming (1998).
- M. BONINO, et al., "Retention of D<sub>2</sub> and DT in Plastic Shell Targets Using Thin Aluminum Layers," Presented at the 11th annual Target Fabrication Specialist Meeting, Orcas Island, Washington (1996).
- E. L. ALFONSO, J. S. JAQUEZ, and A. NIKROO, "Using Mass Spectrometry to Characterize Permeation Half-Life of ICF Targets," *Fusion Sci. Technol.* 49, 773 (2006).
- H. HUANG, R. B. STEPHENS, S. A. EDDINGER, J. GUNTHER, A. NIKROO, K. C. CHEN, and H. W. XU, "Nondestructive Quantitative Dopant Profiling Technique by Contact Radiography," *Fusion Sci. Technol.* 49, 650 (2006).