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BY OVERCOATING WITH GDP**

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ABSTRACT

Free-standing, transparent, two millimeter DT-filled capsules must be fabricated so that ICF scientists can conduct cryogenic layering experiments on NIF-scale targets. To date, cryogenic experiments have used capsules and hemi-shells fitted with fill tubes simply because capsules able to hold over 100 atm of DT gas at room temperature were not available. This report describes one type of transparent capsule that was successfully permeation-filled with 200 atm of helium. It consists of a large glass shell (~1300 o.d. \times 3 μm wall) over-coated with a thick layer (~100 μm) of Glow Discharge Polymer (GDP). We are now filling capsules of this type with over 100 atm of DT to learn if they are suitable for National Ignition Facility (NIF) cryogenic layering experiments.

I. GLASS SHELL SELECTION

The glass shells we selected were made in GA's high temperature (1650°C) vertical drop tower. They had the following characteristics:

1. Wrinkle-free appearance
2. Wall thickness of $\geq 2.5 \mu\text{m}$ (Note: For our drop tower-produced glass shells the best wall uniformity is generally achieved when wall thickness is less than 3 μm .)
3. Diameters greater than 1200 μm
4. Wall Non-Concentricity (NC) $\leq 20\%$

where: $\text{NC} = \epsilon/\text{Avg } W$, ϵ being the distance the center of the spherical shell void is offset from the center of the spherical outer surface.

The glass shells we selected had wall thicknesses of $\sim 3.0 \pm 0.5 \mu\text{m}$. All shells met the required $\leq 20\%$ NC specification and averaged 10% NC. Optically, few shells were found that appeared wrinkle-free. AFM spheremapping of a half dozen shells revealed that their power spectra fell above the NIF capsule standard in modes ~ 10 to 50 (see Fig. 1). We selected the best shells available as determined by optical inspection.

II. GDP-COATING

The glass shells were bounce-GDP-coated in groups of less than 15 shells per batch to minimize shell collisions and thereby produce a better surface finish. (Note: in the future we hope to use a "spinning pan" GDP-coater that will roll shells rather than bouncing them while coating. This gentler motion will improve surface finish and allow us to coat more shells per batch.) A total of 50 capsules were prepared in four batches. After GDP coating, interferometric characterization indicated that the 4π NC of all the coated shells was $\leq 2\%$.

Optical microscopy revealed that many of the GDP-coated shells now looked "wrinkled" around their equators. Apparently, the lens effect of the GDP coating brought out wall distortions that were not readily visible when we optically culled the glass shell batch.

III. BUCKLE TESTING

To determine how much internal gas pressure these GDP/glass capsules could hold, we conducted burst test experiments using helium as the fill gas. Helium has the advantage that it will permeate through glass at least an order of magnitude faster than will hydrogen. Thus, by

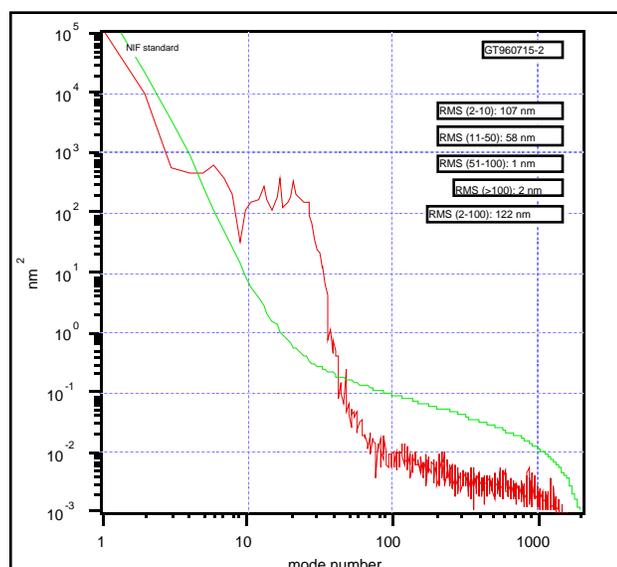


Fig. 1. Power spectrum of typical 1200 x 3 μm drop tower glass shell.

using helium, we could conduct more burst test experiments in the same amount of time.

Before burst testing the capsules, we sought to optimize the fill protocol by first conducting capsule buckling experiments to learn how large the fill steps could be. We used a batch of $\sim 1200 \times 3 \mu\text{m}$ glass shells coated with $\sim 110 \mu\text{m}$ of GDP for the buckle testing. We found that the capsules survived up to 50 atm of buckling pressure at room temperature. We also needed to know if the shells could survive such pressures at the fill temperature required for the helium (and ultimately DT) fill. Furthermore, we wanted to fill the shells at as high a temperature as practical to minimize the fill time (and ultimately DT exposure time at high temperature) yet keep the temperature low enough that the GDP layer would not be adversely affected.

We chose 320°C for our first fill experiment. Based on the apparent strength of the capsules, as indicated by their ability to withstand the room temperature buckle test, we chose to fill the shells with 100 atm of helium using 25 atm steps, holding the shells fifteen minute at each step. We kept the shells at temperature over-night so that the shell fill would come to equilibrium. The shells survived the 100 atm fill, though we noted that the GDP had turned considerably darker on all the capsules. As this filling schedule met our time needs, we began burst test experimentation using the temperature, step size and hold times described above, rather than determining the

ultimate buckle strength of the capsules at elevated fill temperatures.

IV. BURST EXPERIMENTS

For all burst experiments, our fill station setup was such that during shell cool down to room temperature, we had to manually reduce the chamber pressure at a rate which would keep the gas density the same inside and outside the capsule, thereby achieving the desired fill pressure at room temperature. This cool down period was approximately 90 minutes in duration.

We continued our burst test experiments by filling five capsules (three of which had survived the previous 100 atm fill) to 130 atm with helium at 320°C. The two new capsules survived the burst test, however, the three previously filled capsules failed by developing cracks in their glass walls. We suspected glass/GDP delamination due to the high fill temperature. Thus, we decided to next fill the shells at a lower temperature, 280°C, to minimize GDP degradation. We chose this temperature rather than a lower one as a compromise, so not to increase the fill duration to an impracticable degree.

The next fill was run at 280°C, again to 130 atm with helium, but with 20 minutes at each 25 atm step to ensure complete filling. All four new capsules tested survived the fill and had only become slightly darker in color. As a result of this success, we ran all subsequent fills at 280°C.

We next conducted a 160 atm fill using the new lower temperature fill protocol. All ten capsules tested survived this fill including four which had been previously filled.

We then conducted a 200 atm fill (the pressure limit of our fill system). All six capsules tested survived this fill. Rather than using other means to increase the fill system pressure (i.e., a booster pump), we tested capsules with thinner GDP coatings to determine their burst strength.

We conducted a 200 atm fill using capsules that were coated with either 60, 70, 85, 100, or 115 μm of GDP. The 60, 70 and 85 μm -walled capsules burst. In addition, one of the thickest GDP-coated shells failed. The failure of the thick GDP-coated capsule raised the possibility that failure will occur if the glass mandrel has a defect that optical inspection fails to detect. We therefore tested the remaining GDP-coated capsules to “only” 100 or 150 atm to cull out the defective shells. The results of the burst test

experiments are shown in Table I. This table includes the results of a deuterium fill that will be discussed later.

V. GAS FILL CHARACTERIZATION TECHNIQUES

A. Out-gassing in a Closed Volume

To verify that apparently intact capsules were not leaking due to micro-cracks, we assembled a system that used a pressure transducer to measure the rate of pressure rise in a small volume, resulting from out-gassing from a single helium-filled capsule. This system proved effective in verifying capsules fills and half-lives.

B. Weighing

Weighing was the most practical tool for quantifying the helium gas fill and determining room temperature permeation half-lives when processing multiple capsules. However, care had to be taken when using the weighing technique. Shell mass changed as a result of (1) the mass loss of GDP caused by the high temperature fill and (2) oxygen and water vapor uptake by the GDP coating after the fill.

Because of the uncertainty of the mass loss of the

GDP coating during the fill, we chose not weigh the shells before and after the fill to determine the mass of the fill gas. Rather, we determined the fill gas mass by weighing the shells immediately after the fill and again, several days later, when the shell had emptied. We addressed the problem of oxygen and water vapor uptake by storing the capsules in a vacuum desiccator between weighings.

C. Interferometry

We also used interferometry to measure the gas fill and the out-gassing rate. Several factors complicated this approach. Firstly, the index of refraction of helium is very small ($n = 1.000034$) compared to that of hydrogen ($n = 1.00014$). The change in the optical path length through a capsule filled with helium is about one fourth what it would be if the capsule was filled to the same pressure with hydrogen. For the double-pass white light interferometry system used at GA, a 100 atm helium fill of a 1200 μm i.d. shell, would cause an increase of about 5 μm in the optical path length. This would be readily measurable if not for the second problem we faced. The 100 μm thick GDP coating causes significant dispersion in the interferometric fringes making it very difficult to identify the most intense white light reference fringe. Instead of the operator selecting the most intense fringe

Table I
Burst Test Experiments Summary

Fill	Fill Gas & Pressure	Fill Protocol	Results/Observations
1	Helium 100 atm	320°C; 25 atm steps 15 min. per step	6 of 6 shells survived All shells turned dark brown in color
2	Helium 130 atm	320°C; 25 atm steps 15 min. per step	3 previously filled shells failed by glass cracking 2 new shells survived
3	Helium 130 atm	280°C; 25 atm steps 20 min. per step	4 of 4 new shells survived
4	Helium 160 atm	280°C; 25 atm steps 20 min. per step	10 of 10 shells survived 4 shells had been previously filled
5	Helium 200 atm	280°C; 25 atm steps 20 min. per step	6 of 6 shells survived 4 shells had been previously filled
6	Helium 200 atm	280°C; 25 atm steps 20 min. per step	5 of 15 shells survived All but one shell that failed had < 90 μm walls
7	Helium 150 atm	280°C; 25 atm steps 20 min. per step	10 of 11 shells survived (A fill to eliminate defective shells)
8	Deuterium 90 atm	280°C; 20 atm driving pressure; 6 atm steps 2 hours per step	6 of 6 shells survived Shells were slightly darker than those filled with helium in one day
9	Helium 100 atm	280°C; 25 atm steps 20 min. per step	6 of 6 shells survived (A fill to eliminate defective shells)
10	Helium 100 atm	280°C; 25 atm steps 20 min. per step	1 of 3 shells survived (A fill to eliminate defective shells)

from about six visible fringes, now he or she must choose from about 20 or 30 visible fringes. Thus, the accuracy of the measurement was reduced from ± 1 fringe ($\sim 0.3 \mu\text{m}$) to ± 3 or 4 fringes ($\pm 1.0 \mu\text{m}$).

We addressed the dispersion problem by inserting glass coverslips into the reference arm of the interferometer when it was time to identify the reference fringe in the shell. The dispersion caused by the addition of $\sim 400 \mu\text{m}$ of glass effectively canceled out the dispersion caused by the $\sim 100 \mu\text{m}$ thick GDP layer. We finally chose to rely on the simpler weighing technique to verify our helium fills and determine fill half-lives. Interferometry may prove to be the preferred technique when DD or DT fills are considered.

VI. HALF-LIFE RESULTS

The shells tested in this study ranged in size from 1200 to 1500 μm inner diameter and had glass wall thicknesses of $3.0 \pm 0.5 \mu\text{m}$. The average half-life for these shells for helium at room temperature was 10 hours, the standard deviation was 3 hours.

VII. DEUTERIUM FILL TEST

Another issue we addressed in this study was the ability of the shells to withstand a week-long hydrogen isotope fill. We looked at our helium half-life data to find an appropriate fill protocol for deuterium. Based on the helium half-life at room temperature for these shells, we deduced that the glass wall contained $\sim 95\%$ glass formers. Using this factor, we calculated that the shells would have an average deuterium half-life of 4 hours at 280°C .

Using this estimate, we made a conservative deuterium fill protocol at 280°C under the constraint that our fill system had to be manually operated. During the 10 hour workday, we established a 20 atm driving pressure and thereafter took 6 atm steps every 2 hours; the shells came to fill equilibrium overnight; then we repeated the fill protocol over the course of a week. Using this protocol, we filled the shells with 90 atm of deuterium. The shells survived the fill and were only slightly darker in color than those filled with helium in less than 8 hours.

VIII. CRYO TESTING

We were also concerned that the capsules might not survive cryogenic cycling in future DT experiments. To address this issue we tested both helium- and DD-filled shells by immersing them in liquid nitrogen overnight. We "thawed out" the shells in a vacuum desiccator so they

would remain dry. Optical inspection and weighing confirmed that the shells had survived the temperature cycling and had not lost any gas. Subsequent testing verified that the shells' out-gassing rate at room temperature had not been affected by the cryogenic cycling.

IX. SUMMARY

We have fabricated GDP-coated glass capsules that can be filled to over 100 atm with helium or deuterium. We used a fill temperature of 280°C that slightly darkened the GDP layer. Quantifying the opacity of this layer as a function of time at temperature was not addressed in this study. Table II summarizes the results of our burst test experiments.

In the case of helium, we found that shells could be re-filled numerous times. We did not attempt to re-fill deuterium filled shells.

The optical clarity and thus the utility of these composite capsules are most severely compromised by the wrinkled glass walls. The glass shells we used were made from glass frit containing volatile organics that inflate the shells in the high temperature tower. The shells thus produced typically contain a quarter of an atmosphere of residual gases. When the still hot shell falls into the cool zone of the tower, the partial pressure inside the shell coupled with the shell wall surface tension act to make the shell collapse. Unless the shell wall is instantaneously frozen before the collapse begins, it will become wrinkled.

Recent glass shell experiments at GA are addressing this wrinkling problem. Large glass shells having wrinkled walls were filled with several atmospheres of helium. These shells were then individually dropped through a very short (two or three feet) high temperature, vertical tower. Because the shells contained over an atmosphere of gas, the shells increased in diameter when they melted in the tower and they no longer appeared wrinkled. AFM spheremapping of re-blown shells has confirmed that the power spectra in modes under 50 has improved considerably. Though this technique for improving the optical clarity of large shells is simple and looks very promising, several experiments must be conducted to determine whether or not "re-blown" glass shells are suitable for this application.

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