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DISCLAIMER

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New high gain designs for direct drive ignition on NIF require foam shells.[1] Scaled down versions of these designs are needed for near term experiments on the OMEGA laser facility at the Laboratory Laser Energetics (LLE). These shells need to be about 1 mm in diameter and 50–100 μm wall thickness and densities of 100–250 mg/cc. In addition, a full density permeation seal needs to be deposited for retention of the fill gas at room temperature or the ice at cryogenic temperatures. We have fabricated such shells using Resorcinol-formaldehyde (R/F) as the selected foam material due to its transparency in the optical region. Extensive characterization of the wall uniformity of these shells has been performed. The foam shells have ~ 5%–6% non-concentricities on the average. A full density permeation seal has been deposited on the R/F shells using two different techniques. In the first technique R/F shells are coated directly with plasma polymer to thicknesses of 3–4 μm. In the second technique, R/F shells are coated with polyvinylphenol, using a chemical interfacial polymerization technique. Data on surface finish and gas retention for R/F shells coated by both methods are provided.

I. INTRODUCTION

New high gain designs for direct drive ignition on NIF require foam shells. As in the case of thin CH shells designed for NIF and currently fabricated[2] and used at OMEGA for cryogenic shots, initially such foam shells are needed in a scaled down version for shots on OMEGA. In particular, the foam shell diameter needs to be the canonical OMEGA size, which is currently ~ 800–900 μm. The current wall thickness and density requirements depend on the application. The wall thickness for eventual cryogenic shots needs to be in the range of 50–100 μm, while the density is required to be in the relatively wide range of 30–150 mg/cc. The foam shells used as room temperature surrogates need to have wall thicknesses of 80–120 μm and densities similar to

that of D_2 or DT ice, ~ 180–250 mg/cc. Since simulations have found that the implosion was insensitive to as much as 30 atom % oxygen, the foam material could be an oxygen containing CH based foam as described below. In addition, a full density gas barrier, ~1–5 μm in thickness, is needed on the outside of the foam layer to prevent evaporation of the fuel ice in the case of cryogenic experiments or to hold the gas fill in case of room temperature shots.

This paper discusses the fabrication, characterization and testing of such overcoated foam shells. Since previous workers[3–6] had determined the required conditions for the R/F foam fabrication process, we will only briefly discuss R/F foam shell fabrication, pointing out some major differences we have observed in our work compared to the previous work. Also we will provide a summary of the extensive data obtained on the quality of the shells produced which has been absent in most previous reports. We will also discuss the two different overcoating processes we have used. These overcoatings had been also used previously on R/F foam shells.[7,8] In this paper we have examined and report on their physical properties of interest which are important for fielding coated foam targets. They include surface smoothness, gas retention, and strength of the coated foam shells.

II. FABRICATION OVERVIEW

The fabrication effort involved three separate tasks. Our fabrication strategy was geared towards obtaining a product that could be fabricated and used with reliability in a relatively short period of time. For this, given the previous body of work on various foam systems, we concentrated on fabricating foam shells using the known chemistries rather than developing new foam systems. Key aspects of the fabrication were the quality of shells and the ability to produce large quantities of high quality shells.

An important second step beyond shell fabrication was deposition of an appropriate full density gas barrier.

This barrier needed to be uniform in thickness and be gas retentive. While the production of a gas retentive barrier was developed previously,[2] the reliability of this step was very much in question. This is especially true for R/F foam shells. In particular, there was very little data on gas retentiveness of the various seal coats attempted previously.

Full characterization of the foam shells and the gas barrier was the important third task for this effort. A key requirement of the fabrication process was rapid characterization of a large quantity of shells which is needed for obtaining proper statistics on foam shell batches. This required optical transparency of the foam shells to allow routine optical measurement techniques such as white light interferometry to be used. This allowed important shell metrics such as shell wall thickness uniformity to be measured.

III. FOAM SHELL FABRICATION

We initially attempted fabrication of foam shells involving an oil based solvent, since this would involve simple and direct extension of our full density poly-alpha-methylstyrene (PAMS) shell fabrication process.[9] Here the foam material is dissolved in an oil-like solvent and is formed using the droplet generator and stabilized between two water phases. This is the typical W/O/W (water-oil-water) microencapsulation system. Fabrication of these types of foam shells involved minimal changes to the droplet generator setup and was readily accomplished. We examined fabrication of foam shells previously used in the ICF area, including TMPT, EGDM, and subsequently DVB which was at the time concurrently under development for IFE.[10] For R/F which is water soluble, the encapsulation system needed to be reversed and was very much unlike the PAMS fabrication process. This reversed encapsulation system presented several challenges which were addressed in the previous work.[6]

Initially, foam systems developed at Japan's Institute of Laser Engineering (ILE) were investigated as these had yielded shells which had been used for cryogenic work. These included TMPT and EGDM at 100 mg/cc both of which are CH based foams which contain over 10% oxygen. They have pore sizes in the sub-micron to a few micron range, scattering visible light too much to be generally transparent. While the foam shells reported in the Japanese work were sufficiently transparent due to their relatively smaller wall thickness (~ 10 µm) and diameter (~ 500 µm), the thicker, larger shells made by us in this new required size range were far from transparent once dried. The opacity leads to difficulty in applying standard optical characterization techniques for measurement of these shells. In addition and probably more importantly, this would not allow the eventual required characterization of the ice layer during cryogenic layering of ice carried out at OMEGA.

We also fabricated DVB (divinylbenzene) shells at 100 mg/cc density. DVB shells are attractive in the IFE area because they have the distinction that they are pure CH and do not contain oxygen. However, as in the case of TMPT and EGDM, DVB foam, due to its large pore size, is too opaque for visible light characterization. Nonetheless, x-ray radiography was used to determine wall thickness and uniformity of a small subset of DVB shells (Fig. 1). The results showed that DVB shells with good sphericity (< few microns) could be made which had good wall uniformity (< 10%). But the characterization was too time consuming for this foam system to be considered a viable option for this application. In addition, another observation was that the dried DVB shells were brittle and would crack easily during handling.

R/F foam is transparent optically due to its small pore size of <100 nm. It therefore became the natural choice for our application. R/F is also CH based with an oxygen content to be 14 atom % measured by combustion analysis of a representative sample. This was well within the acceptable oxygen content for the LLE application as mentioned before. Fabrication of R/F shells is more complicated when compared to W/O/W systems since the R/F shells are made employing a reverse O/W/O (oil-water-oil) system which can be a more difficult working system.

Lambert and coworkers had determined optimum conditions for fabrication of low density (50–100 mg/cc) R/F foam shells.[6] One of the key findings in that and previous work was the apparent need for processing the foam material so that it gelled very quickly (~ 15 min) after droplet generation. This was needed to ensure the survival of the wet foam preform. Also, a propeller system was used during the curing process for shell agitation, resulting in survival of only 1/3 to 2/3 of shells. While we used similar recipes to these previous efforts for for the ~ 100 mg/cc R/F foam shells, our observations were not
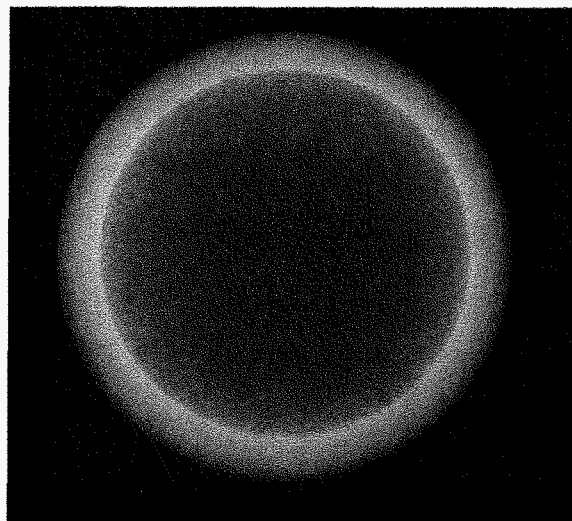

Fig. 1. X-ray radiograph image of a DVB shell, ~ 2 mm diameter and ~ 150 µm thick. DVB shells are opaque optically due to their large cell size and need to be characterized by x-rays.

exactly the same. The differences will be detailed in a future publication but we mention the most striking difference briefly here. We found that fast gelation was not essential for perform stability. In fact, too fast a gelation resulted in poor wall uniformity for lower density shells, ~ 100 mg/cc, and that it was detrimental to shell fabrication for the higher densities of ~ 200 mg/cc. For these higher densities major modification of the previous recipes needed to be made in order to produce shells. Also, we used a rotary flask system, similar to that currently used in the PAMS process,[11] for shell agitation and obtained yields of nearly 100% consistently allowing production of thousands of shells per batch. Figure 2 shows an example of a portion of a batch of R/F foam shells in isopropanol prior to the critical point drying step.

We conducted extensive optical characterization of dried R/F shells. The characterization involved measurements of shell metrics similar to what is done on PAMS shells. Figure 3 shows an example the statistical sampling performed on several batches. Many batches were examined in this way with similar results at the 100 mg/cc density. Approximately 20%–25% of shells had delta walls of <5 μm which was adopted as an initial standard for wall thickness uniformity.
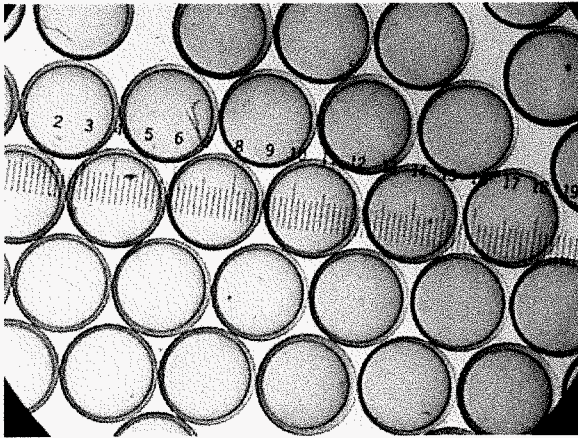

Fig.2. Sample of a batch of R/F shells made by microencapsulation.

IV. OVERCOATING

A key requirement of the full density overcoating is that it serves as a gas permeation barrier. When we began our overcoating work, the only proven successful technique for overcoating foam shells which had been demonstrated to produce a gas permeation barrier was the chemical process of interfacial polymerization of polyvinylphenol (PVP) developed by Takagi et al. at ILE.[3] This had been done on TMPT and EGDM foams but not R/F. Therefore, we attempted PVP coating on R/F shells, however, because of the relatively small pore size of R/F foam we also conjectured that it might be possible to close the foam pores by applying a coating in a plasma process such as glow discharge polymer (GDP) process.
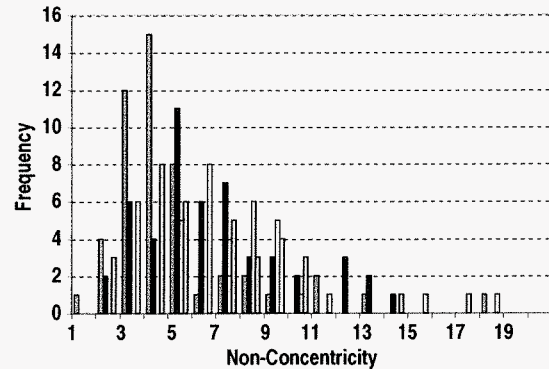

Fig. 3. Histograms wall thickness uniformity obtained on several batches. Typically over 50 shells were examined from each batch to obtain proper statistics. ~ 20%–25% of shells had wall thickness uniformity below the required ~ 5 μm.

GDP had been deposited on R/F foam shells previously, but there was no direct data on gas retentiveness of such coatings, especially at the 1–5 μm thickness required for the LLE experiments. The key issue was then to examine gas retention by the two different coating techniques. Other issues with the different coatings were surface finish and strength. The surface finish obviously impacts target performance during the implosion and needs to be below ~ 50 nm. The coating strength is important because the coating needs to hold the required gas fill at room temperature. Proper strength is crucial for allowing rapid filling of shells to very high pressures (~ 1000 atm) in the cryogenic application.

We were able to deposit thin, ~ 3 μm, GDP coatings on the R/F shells and found that it was indeed sufficient to close the pores in the R/F foam to form a gas permeation barrier, but not in all cases. In the GDP process R/F foam shells were agitated using the standard piezo-electric shaking mechanism used to coat full density shells. It was found that the foams shells do not "bounce" very well initially during the coating process and a much larger (~ 2x) than usual power level to the piezo electric tube was required. However, as more and more GDP was deposited on the shells they appeared to move more freely and eventually behaved as the full density shells did during the coating. Given the different agitation behavior of the R/F foam shells there was initially concern regarding the uniformity of the coatings. Interferometric measurements did not on average indicate any gross thickness uniformity of the GDP overcoating. Nonetheless, a number of individual shells were examined using the wallmapping process,[12] which could map the thickness of the coating around the shells as a function of angle in several orthogonal directions. Figure 4 shows one such trace indicating that despite the anomalous agitation behavior of R/F foam shells the GDP thickness uniformity on these shells was very good (< few tenth of micron), similar to that obtained on full density shells. Also, we measured no significant diameter or wall thickness shrinkage of the
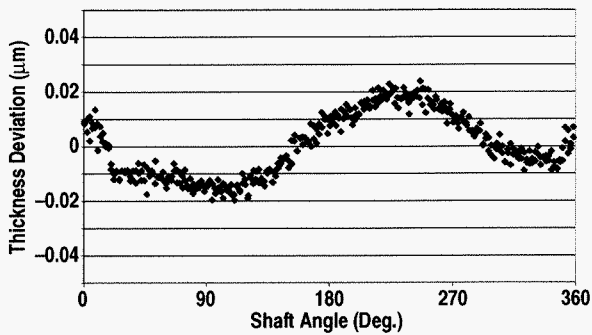
Fig. 4. Wallmap of the GDP coating deposited on a ~ 100 mg/cc R/F foam shell. The GDP uniformity is very good similar to what is obtained on full density shells.

foam shells after the GDP coating process, which indicated a low level of stress in the coatings and no significant density change in the foam layer. It should be mentioned that DVB shells were also coated with GDP but even after deposition of nearly twenty microns of coating a contiguous layer was not observed.

PVP coatings were successfully deposited on R/F shells in our work as well (Fig. 5). The details of the deposition process will be presented in a future publication. The PVP coatings were also examined for uniformity and were found to be similar to GDP coatings. However, a troubling feature of the PVP coatings was the large amount of shrinkage observed in the foam shell diameter (Fig. 6) and some increase in the foam shell wall thickness after critical point drying of the PVP coated
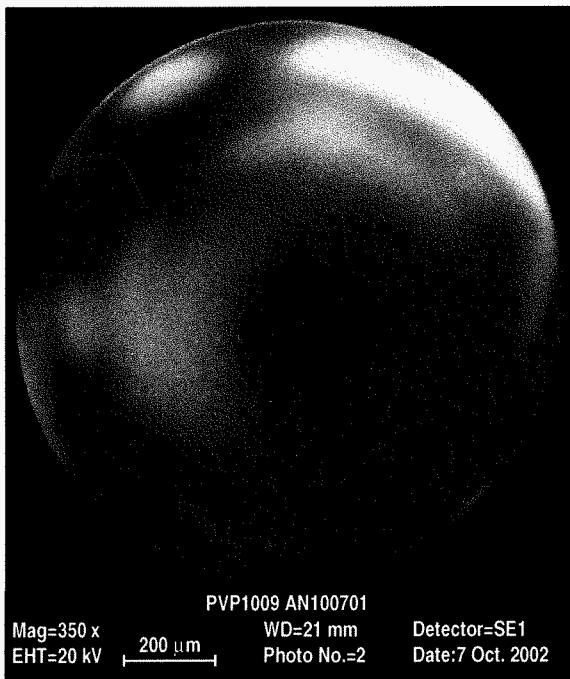

Fig. 5. SEM picture of PVP coated R/F foam shell. Proper processing led to depostion of uniform coatings that did not delaminate after drying.
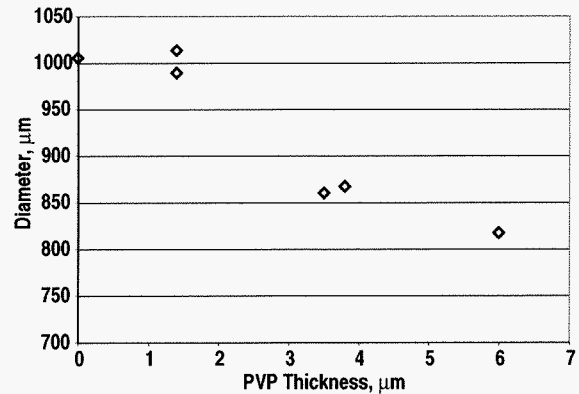

Fig. 6. Thicker PVP coatings led to significant shrinkage of the foam shell diameter. The commensurate wall thickness increase was about the same as the diameter shrinkage, percentage wise, indicating a lsmaller volume for the foam and hence densification of the foam shell.

shells. Overall, the foam layer volume was smaller than prior to the coating. This diameter shrinkage was dependent on the PVP thickness, being nearly 20% for thicknesses of ~ 3 μm. This obviously meant that the foam layer had densified, but this densification was not necessarily uniform and was hard to quantify. Only very thin coatings of < 1 μm resulted in negligible shrinkage of the foam layer. This observed result with PVP coatings was very troubling and indeed as discussed later was the main reason why GDP coatings were eventually chosen as the preferred seal coat for R/F foam shells.

The gas retention of the GDP and PVP coated shells was then examined by first permeation filling of shells with argon and then observing the argon content of shells as a function of time by tracking the argon x-ray microfluorescence (XRF) signal from the shells. Figure 7 shows the typical signal from those GDP coated shells that were gas retentive. The rate of argon loss from these shells was very similar to that observed in thin wall, ~ 3 μm, full
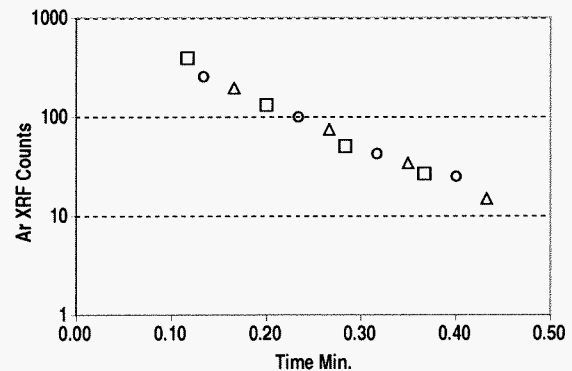

Fig. 7. Decay of argon signal from three different argon filled R/F shells coated with ~ 3 μm of GDP. This rate of decay was similar to what is observed on pure GDP shells, indicating gas retentiveness of the GDP coated foam shells.

density CH shells made using the GDP process. Unfortunately, not all shells in a coating batch displayed the same behavior with some not holding gas. Over 100 GDP coated shells were tested in this way to obtain reliable statistics for the fraction of shells that hold gas. In nearly 30%–35% of the shells the argon signal was completely absent, indicating that the GDP coating had not completely covered the foam pores and the argon inside the shells had escaped before the shells could be examined by XRF. PVP coated shells were examined the same way and it was found, rather surprisingly, that 10% of PVP coated shells did not hold gas either. In most cases, for GDP and PVP, optical examination of shells that failed the gas retention test did not reveal any obvious flaws in the shells. Therefore, the exact reason for lack of proper sealing of the R/F foam in those shells is unknown. This pointed out the necessity of testing every single shell, either GDP or PVP coated, for gas retention before it could be delivered to LLE for laser shots or offline cryogenic experiments.

The surface finish of PVP shells had been reported to be as low as 10 nm RMS when deposited on TMPT shells when examined by phase shift interferometry (PSI) in the ILE work.[3] The PVP coatings, ~ 3 μm, we deposited on R/F shells exhibited a larger amount of roughness as examined by both PSI and atomic force microscopy (AFM). The AFM results were generally higher, ~ 40 nm RMS on a 20×20 μm patch, compared to the PSI measurements which yielded ~ 16 nm RMS for the same shell. In each case several different areas were sampled to ensure proper sampling of an admittedly nonhomogeneous surface. But the results were similar with the AFM roughness numbers being consistently ~ 2 times higher. The origin of this difference is probably related to the lower resolution of the PSI technique, but detailed explanation of this is beyond the scope of this paper. The main result is that the roughness of the PVP coated R/F shells was only slightly higher than what was reported previously when examined using the same technique (PSI), but higher using the current accepted roughness measurement technique (AFM). The surface finish of GDP coated shells depended on the coating pressure used. At the standard pressure of ~ 75 mTorr the AFM measurements indicated a roughness of ~ 80 nm, while when the coating pressure was increased to 500 mTorr, the roughness dropped to ~ 40 nm. A hybrid of the two, with early high pressure coating followed by lower pressure coating also produced coatings with the lower roughness values. In fact, for a direct comparison some GDP and PVP coating shells were AFM sphere-mapped. The results are shown in Fig. 8. Surprisingly, the GDP coated shells were actually smoother in the high modes than the PVP coated shells, although there was some variation for the GDP coated shells. The basic conclusion is that the GDP coatings are actually not any rougher than PVP coatings, at least on R/F foam shells.

Finally, we buckle tested some ~ 3 μm thick PVP and GDP coated shells. Nearly 40 shells were tested of each
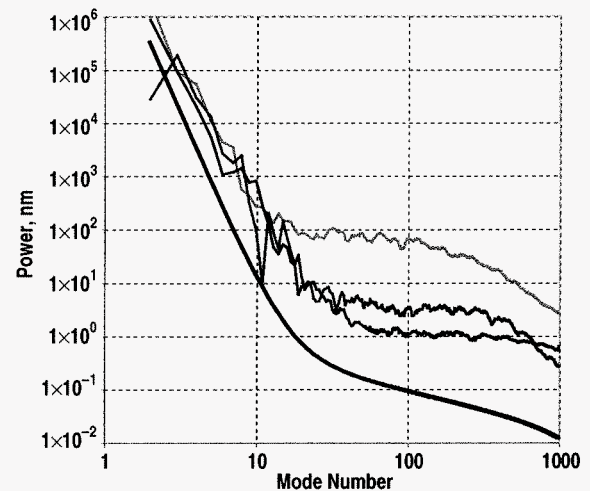

Fig. 8. AFM power spectra of R/F shells coated with PVP and GDP coated shells. The trace with the largest high mode roughness is that of a PVP coated shell. The two with the lower high mode roughness are those of two different GDP shells. GDP coatings on R/F shells were as smooth or smoother than PVP coated shells.

type. The PVP coated shells were found to have the highest relative strength when the slight dimensional differences (thickness and diameter) are accounted for. The PVP coated shells were 1.5× stronger in buckling than pure CH shells, while the GDP coated shells were only 1.2× stronger. Therefore, it appears that the overcoated foam shells are at least as strong as the pure CH shells. Therefore, it should be possible to fill them sufficiently fast to 1000 atm of D_2 pressure required for cryogenic shots. A GDP coated foam shell was filled and cooled cryogenically at LLE.[11]

V. CONCLUSION

R/F foam shells ~ 100 mg/cc have been fabricated for use at LLE as the scaled down version of new high gain designs for direct drive ignition on NIF. These shells are ~ 800–900 μm in diameter and 50–100 μm in wall thickness. The batch wall uniformity of these shells has been shown to be sufficiently good, with nearly 25% of shells having delta walls of < 5 μm as desired. Gas retentive, full density coatings were deposited using both GDP and PVP on R/F shells. Both coatings were shown to be gas retentive, although not at 100%. This necessitates testing of all shells for gas retention prior to use as laser targets. The surface roughness of the two types of coatings is similar and is low enough (~< 50 nm) for use as laser targets. The buckle strength of coated R/F shells was measured for both types of coatings. The buckle strengths are about 50% higher than pure "standard" CH shells and should be sufficient to allow high pressure filling and then cooling to cryogenic temperatures. A GDP coated R/F shell was filled in this manner and an ice layer was formed

inside it. Higher density ~ 200 mg/cc shells have also been fabricated for use as room temperature surrogates for the solid fuel, but the R/F fabrication process needs to be optimized to produce higher percentages of shells with uniform wall.

ACKNOWLEGMENT

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