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Prospects for 2 mm Diameter NIF Polymer Capsules*

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Abstract — The National Ignition Facility (NIF) will require 2 mm diameter shells with a flexible composition profile and variable thickness. These shells must meet out of roundness (oor) and surface roughness specifications which are at least as good as the best 1/2 mm shells shot on Nova. These perturbations are generally expected to become worse with increasing diameter, so there is some concern about the prospects of meeting those exacting specifications with shells with four times larger diameters.

The PAMS/GDP technique we are currently using for 1 mm diameter OMEGA shells seems capable of producing satisfactory NIF shells. It can produce shells with a wide variety of dopant elements and concentration profiles, and wall thicknesses.

Comparing the oor of NOVA and OMEGA shells made with the PAMS/GDP process with that of larger shells made of polystyrene by a similar process at the Osoka University Institute of Laser Engineering (ILE) suggests that the perturbations during curing can be less important than previously believed. We see no fundamental barrier to providing shells to NIF specifications. However, process optimization will be required.

I. INTRODUCTION

As fusion drivers have grown in power, the targets they can implode have grown in size. The initial targets in the 1970's were less than 100 μm in diameter. In the late 1980's targets grew by several hundred microns for NOVA experiments. Now, in the late 1990's, OMEGA requires 1 mm diameter targets. In five years, we will have to deliver 2 mm shells to NIF. Along the way, the specifications for these targets have grown tighter—The NIF target specifications are derived from the best shells used on NOVA. In order to accommodate the improved specifications, the process by which the shells have been made have evolved along with the size. Initially hollow glass shells, used for reflective paint, were bought in bulk and sorted. As specifications grew tighter and sizes increased, a drop tower technique was developed, and then a microencapsulation technique which produced rounder and larger shells. A decomposable mandrel technique being developed now is the most promising method for making polymer shells for NIF. Its attributes will be described in Section II. Unfortunately, the spheridizing forces decrease, and the perturbing forces increase as the shells increase in size. There is a real concern about how spherical a shell we can make at the NIF size. This has led to various investigations into the real limitations in production of smooth spherical shells. Cook et al [1] previously discussed forces which would limit the roundness of a sphere, and concluded that there might be problems attaining the NIF goals. We show in the Section III that recent data from ILE and from GA suggest that the problems pose less severe limitations than had been suggested and that the NIF goal is attainable.

II. GDP/PAMS NIF SHELL CHAR

A new technique invented by Letts [2] has become the most promising method for making polymer shell targets for NIF. It is currently the preferred technique for making polymer capsules with 1 mm and below diameters. In this technique, called the depolymerizable mandrel technique or GDP/PAMS, a shell is made from poly (α-methylstyrene) [PAMS] by microencapsulation and then coated with GDP (a polymer made by passing *e.g.* trans-2-butane through a glow discharge). By adding carriers to the gas stream during the coating process, the GDP can be doped with a number of dopants (in our current work we have added Cl, Ti, Ge, Si, and D), at arbitrary radii in controlled compositions and layer thicknesses. The coated shell is heated to break down PAMS into its monomer which permeates through the wall leaving a GDP shell. The surface finish of the pyrolyzed GDP shell is approximately the same as the bare PAMS mandrel (Fig. 1), but the shell thickness and composition are freed from the constraints of forming the mandrel so any desired wall thickness and composition profile can be made. (Fig. 2 shows a 1 mm diameter shell with 0.8 μm thick wall.) Thus, the key obstacle for extending the technique to 2 mm NIF polymer shell targets is making a 2 mm bare PAMS shell with adequate sphericity.

III. SHELL SPHERICITY VS SIZE

The distortion of ICF shells is described by the averaged power spectrum of a series of orthogonal radial traces around the shell (Fig. 3). The lowest order, and experimentally largest amplitude distortion is elliptical and is the one of primary concern here. This distortion is referred to as 'out of round' (oor) and is measured by the

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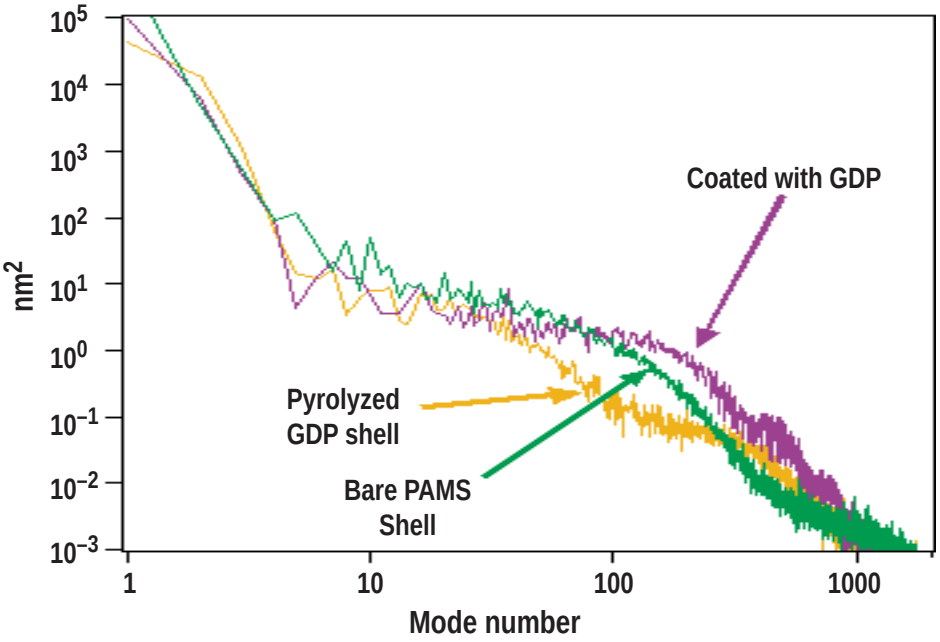

Fig. 1. Power spectra of shells at various steps in the GDP/PAMS process. The green curve is a 1 mm bare PAMS shell ready to be coated. The purple curve is a PAMS shell coated with 12 μm GDP. The orange curve is a GDP shell after the PAMS was pyrolyzed away. The "coated" and "pyrolyzed" shell are from the same shell. The "bare PAMS shell" is from the PAMS shell batch used for the coated and pyrolyzed shell. The differences between the three traces are not significant, but are related to differences in the orientation of the shell when characterized for the power spectra.

difference between maximum and minimum radius. Cook *et al.* [1] developed the following relations to describe oor limitations caused by gravity and by shear when making shells from a plastic containing oil drop in water:

$$\text{oor}_{\text{sag}} = \Delta\rho g r^3 / \gamma \qquad (1)$$

$$\text{oor}_{\text{shear}} = 4\mu G r^2 / \gamma \qquad (2)$$

where r = shell radius, γ = oil/water interfacial tension, g = gravitational acceleration, Δρ = oil/water density difference, μ = water viscosity, and G = linear velocity gradient.

They predict oor to increase as r^3 and r^2 respectively. They are plotted in Fig. 4 using values suggested by Cook et al [1]: γ = 35 dyne/cm, Δρ = 0.02 gm/cc, μ = 10 cp, G = 6 s^{-1}. The plot also contains batch (•) and individual (O) oor values from GA and LLNL measurements of 2, 5 and 7 mm diameter polystyrene (PS) shells made by ILE In 1995, 1/2 and 1 mm PAMS shells routinely made at GA, and a few larger PAMS shell batches made as trials. There is a large scatter in the experimental oor—clearly it is possible to make poor shells. However, one can see that for the best shells the oor of these shell batches is not increasing nearly as fast as described by the shear and sag equations. The alignment of the model curves to experimental oor could well be coincidental; there is substantial uncertainty in the parameters which should be used in them. However, it is clear that for the ILE shells, when corrected for size (using Eqns. 1 or 2), the oor distortion from shear or sag is at least an order of magnitude smaller than the oor of the GA shells.

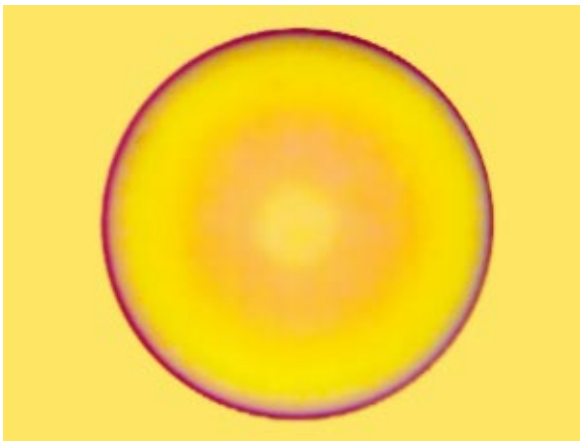

Fig. 2. Shell made from the GDP/PAMS process, ~1 mm diameter, with walls 0.8 μm thick.

To meet NIF specifications, it will be necessary to make PAMS shells that meet or beat the PS oor. We will be looking into the differences between the PAMS and PS processes to learn how to optimize the PAMS process to achieve PS oor's.

IV. SUMMARY

The GDP/PAMS process—GDP overcoating and then burnout of a PAMS mandrel—allows arbitrary wall

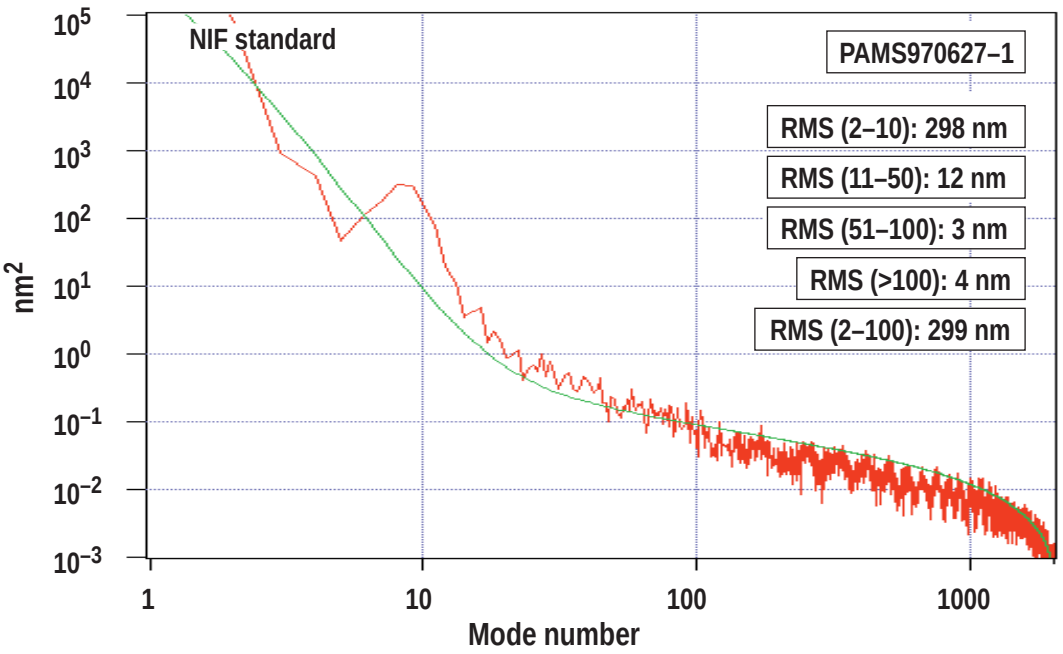

Fig. 3. Power spectrum of a 1.4 mm diameter PAMS shell which had a 4 μm oor (red) and the specification of a NIF baseline shell. Mode 2 corresponds to an elliptical distortion of a shell, and tends to get worse as shells get larger. Modes above ~100 correspond to surface roughness whose amplitude as a function of wavelength is independent of shell size. Since mode numbers are shell circumference divided by distortion wavelength, this part of the curve moves to the right as the shell gets larger; for a 2 mm shell, the experimental and specification curves would overlap.

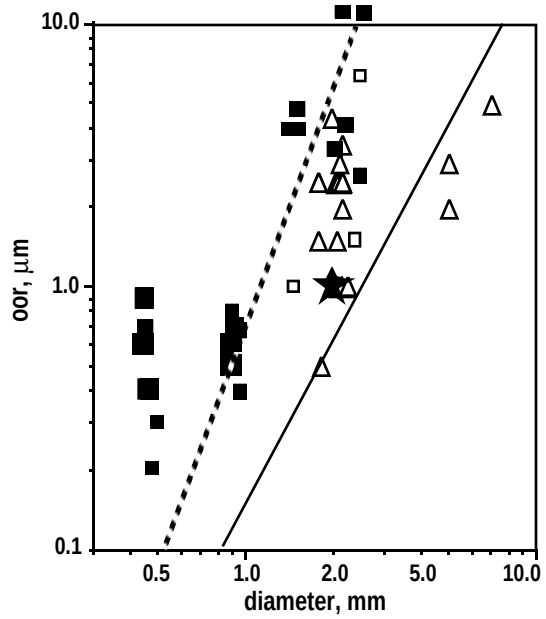

Fig. 4. Shell oor (max-min radius) as predicted by models developed by Cook [1] for shear and sag. In both cases parametric values to use are uncertain; the important fact is that oor is predicted to increase as r^3 in the sag case, and r^2 in the shear case. The squares are experimental data from batches of shells made of poly (alpha methyl styrene) (PAMS) at GA. The solid points represent batch average oor. The open points are measurements of individual shells The triangles are data from measurements by GA and LLNL [3] of poly (styrene) (PS) shells made at ILE (Japan). The oor for dia ~ 2 mm are individual values typical of the shells examined. Those for larger shells are individual values for selected shells. The star the expected NIF oor.

thicknesses and dopant profiles which are necessary for NIF experiments. The surface finish of the final shell is similar to that of the PAMS mandrel.

The primary barrier to shells meeting NIF specifications is the oor of the PAMS mandrels; trial batches of shells of that size have several times the desired oor, as predicted by models of the distortion process. The oor is lower than predicted by these models, and close to NIF requirements for PS shells – a very similar system. Understanding the differences in these processes should give us the ability to produce shells with satisfactory oor.

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