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INVESTIGATION OF LARGER POLY(α-METHYLSTYRENE) MANDRELS FOR HIGH GAIN DESIGNS USING MICROENCAPSULATION

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In recent years we have demonstrated that 2-mm-diameter poly(α-methylstyrene) mandrels meeting indirect drive NIF surface symmetry specifications can be produced using microencapsulation methods. Recently higher gain target designs have been introduced that rely on frequency doubled (green) laser energy and require capsules up to 4 mm in diameter, nominally meeting the same surface finish and symmetry requirements as the existing 2-mm-diameter capsule designs. Direct drive on the NIF also requires larger capsules. In order to evaluate whether the current microencapsulation-based mandrel fabrication techniques will adequately scale to these larger capsules, we have explored extending the techniques to 4-mm-diameter capsules. We find that microencapsulated shells meeting NIF symmetry specifications can be produced, the processing changes necessary to accomplish this are presented here.

I. INTRODUCTION

Indirect drive implosions using 2ω light or direct drive implosions on the NIF will require 3-5 mm diameter capsules.[1] The extension of the 2-mm-diameter work is not necessarily straightforward, thus we have explored the fabrication of larger mandrels using the microencapsulation technique. All potential targets except machined beryllium[2] depend upon this mandrel on which plastic or sputtered beryllium is applied.[3] Thus the mandrel must at least meet the surface finish requirements and symmetry of the final target.

The mandrels are produced by a two-step process.[4] In the first step a thin-walled poly(α-methylstyrene) (PαMS) shell is produced using microencapsulation techniques.[5,6] This shell is overcoated with 10 to 15 μm of glow discharge polymer[7] (GDP) and then pyrolyzed at 300 °C. This pyrolysis causes the PαMS to depolymerize to gas phase monomer that diffuses away through the more thermally stable plasma polymer shell, which retains all the symmetry of the original PαMS shell. Thus our challenge has been to produce larger diameter PαMS shells that meet or exceed design specifications to serve as these initial decomposable mandrels. Though the feasibility of meeting these design goals has been established, the mid- and high-mode roughness does not yet meet design specifications consistently.

II. MATERIALS AND PROCEDURES

II.A. Solutions

Fluorobenzene was freshly distilled at 85 °C. A PαMS (M_w = 400K, Scientific Polymer Products) solution in toluene is water washed multiple times to remove ppm levels of lithium impurities, reprecipitated, dried, and stored under vacuum below -20 °C. The vacuum cold storage seems to reduce a slow air oxidation of the dry PαMS. A solution of 18 wt% PαMS in fluorobenzene (O) is filtered at 0.45 μm, and refrigerated until used, normally within a week. We have found the water washing and the prompt use of the O solution both seem to reduce the presence of vacuoles. A 0.05 wt% poly(acrylic acid) (PAA) solution (M_w = 1000K) is pressure filtered at 8 and 3 μm, yielding a solution with viscosity between 12.8 and 13.3 cP at room temperature. This filtration removes many, but not all PAA gel particles. The PAA solution serves as the initial exterior suspension fluid (W2). A 2 wt% poly(vinyl alcohol) (PVA) solution is vacuum filtered at 0.2 μm and used for exchange as described below.

II.B. Methods

4 mm shells are made by microencapsulation at a rate of about 1 per second, there are from 900 to 1250 shells per rotary flask batch, and the flask is gently rotated horizontally at 30 rpm. A gas flow of 52 cc/min passes through a bubbler filled with fluorobenzene to the top of the W2 solution to slow the curing rate.[8]

After curing, the shells are transferred to a one liter beaker. They are washed at least 10 times with deionized water. The thoroughly washed shells are then placed with 100 ml of water in a small glass bottle. We then add 5 ml of isopropyl alcohol (IPA), and after mixing, the bottle with shells is placed in an ultrasonic bath for 2-3 s, and then allowed to sit overnight. This process is repeated daily to nucleate a small air bubble in the core water of each shell, at which time the shell will float. Typically a total of 60-80 ml IPA is added over 4-6 weeks until the good shells float to the surface and broken shells remain on the bottom. The good shells are then moved into a vacuum oven for 3-4 weeks at 30-35 °C to remove all the core water.

III. INVESTIGATION OF METHODS TO DECREASE MID-MODE AND HIGH-MODE AMPLITUDES.

A summary of 36 runs of 4 mm OD shells is shown in Table I. For all these shells, the nominal diameter is 4050 microns. The out-of-round (OOR - mode 2) is typically 1-3 µm, slightly higher than the desired 1 µm typically seen for 2-mm shells.[6] In evaluating these batches on the spheremapper, the amplitude of the mid-modes (11-100) and high-modes (101-1000) was greater than the desired NIF specification. If the origin of the mid-mode amplitude was Marangoni convection, we anticipated that by slowing the curing rate we might suppress the mid-mode amplitude.[9] We sought to slow the curing by increasing the amount of fluorobenzene in the bubbler, from 60 g to 120 g. The high-mode amplitude presumably has its origins in PAA gel particles that adhere to the curing shell. We knew that PVA seemed to reduce this roughness in smaller shells, so we attempted two different PVA additions. In one method, shown in Figure 1, the PAA solution is removed entirely at about 72 hours, the almost cured shells washed with deionized water, and then the shells are placed in PVA solution. This complete *exchange* of PAA with PVA is method A as listed in Table I. In the second *combination* method (methods B-D in Table I, see Figure 2), different amounts of PVA are added to the PAA solution after curing for 72 h, anticipating that the PVA would remove the particulates from the surface, even though PAA is still present. 24 to 48 h after this initial PVA addition, the entire PAA/PVA solution is removed and replaced with 2% PVA. The shells are then rotated in this 2% PVA bath for another 24 h before the final washing and drying.

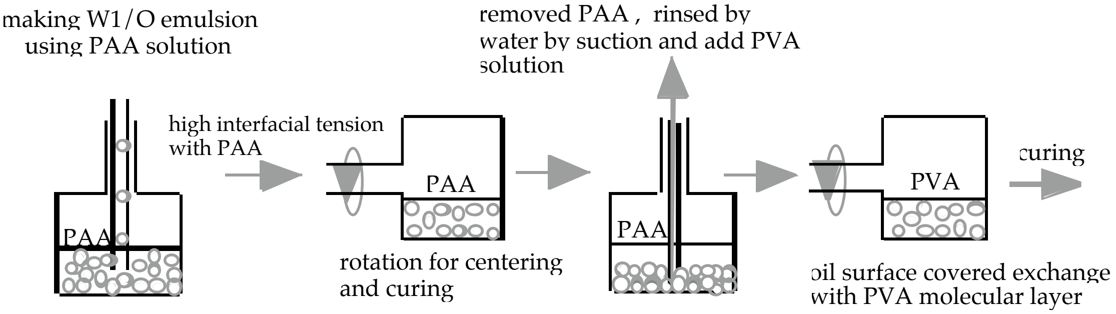

Figure 1. The PVA exchange method (A in Table I).

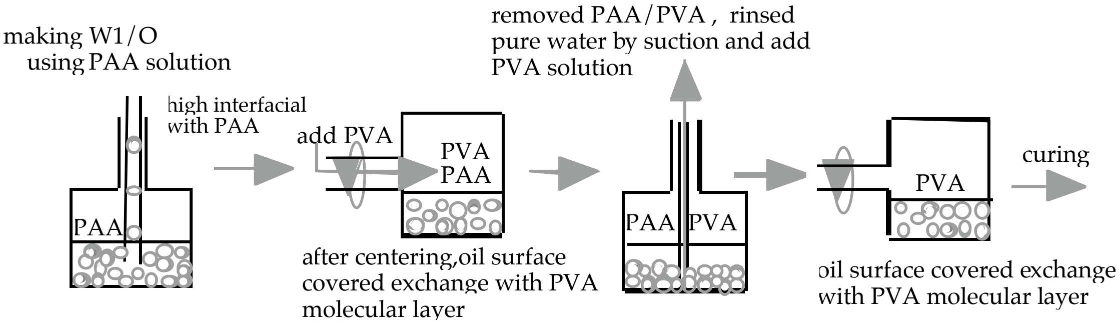

Figure 2. The PVA combination method (B-D in Table I).

Figure 3 shows a plot of spheremapper rms of the amplitudes, across the mid-mode (11-100, open symbols) and high-modes (modes 101-1000, filled symbols), as a function of the amount of fluorobenzene in the bubblers and the type of PVA washing that was done (batch number, see Table I). The graph shows no clear effect of these process variables on the shell surface quality. Either these two variables have little effect, or an unidentified factor is dominating the surface quality.

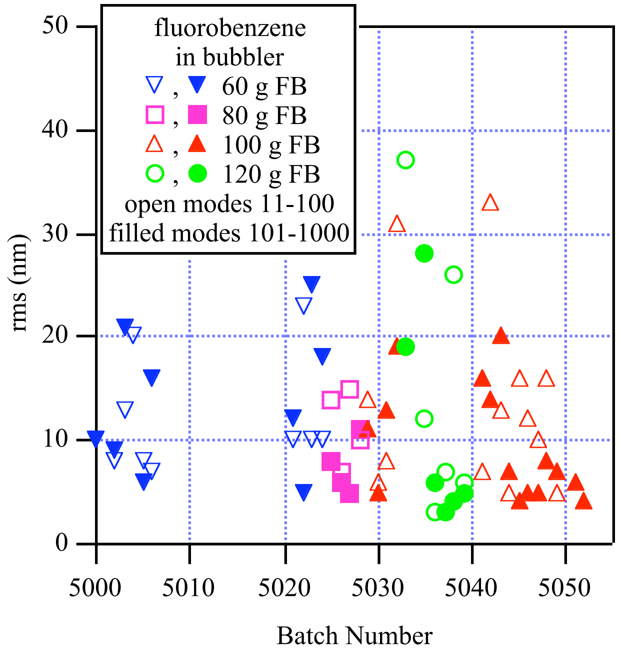

Figure 3. Mid- and high-mode rms of shells by batch. See Table I.

Figure 4 shows the comparison of the mid-mode and high-mode rms, as a function of process variable. Batch 5036 is the leftmost point. Figure 4 clearly shows that while we can get the high-mode rms below 10 nm (and many cases closer to 5 nm), it is the mid-mode amplitude, which is not under clear control, though there is a weak indication that more fluorobenzene in the bubbler, *i.e.* slower curing times, helps. Very few of the batches have a mid-mode <10 nm, and 10 nm is not good enough to be entirely below the NIF curve.

Figure 5 shows three examples of the data for shells. Batch 5036 is clearly the best, with power at the NIF specification curve across virtually all modes.

IV. DISCUSSION AND SUMMARY

The fundamental shell symmetry, primarily the mode-2 OOR and NC, can be fixed by an initial formation and cure in PAA until the oil phase is viscous enough to resist distortion.[6,8] PVA is then effective at removing adsorbed PAA gel from the surfaces of PαMS shells. In addition, increasing the amount of fluorobenzene in the bubbler, and slowing the extraction, seems to improve the mid-mode roughness, but not yet sufficiently to be totally convincing. One possibility for future work is not only to increase the amount of fluorobenzene in the bubbler, but also to increase the time to PVA exchange/washing from 72 hours to some longer time. We have seen that the bubblers lose fluorobenzene at a rate of about 1 g/hour. In the data presented here, we always added PVA at the 72 hour mark, thus with larger amounts of fluorobenzene in the bubbler the shells may not have been sufficiently cured and mid-mode deformations may have formed.

In summary we have improved the production process for PαMS mandrels by utilizing a PAA/PVA combination or exchange process. We cannot yet be sure which PVA method is best. The advantages of PAA for low-mode sphericity are maintained. There exists 4 mm shells that satisfy the NIF specification curve, but we have not yet identified all the process parameters to make such shells reproducibly.

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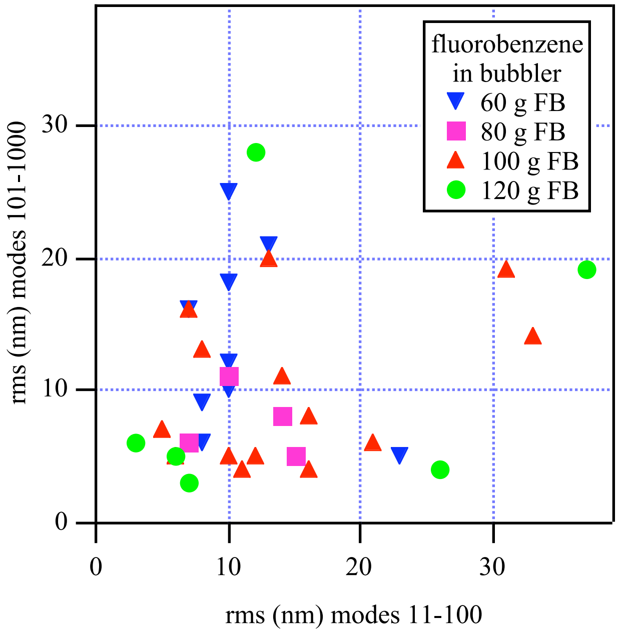

Figure 4. Correlation between mid- and high-mode roughnesses.

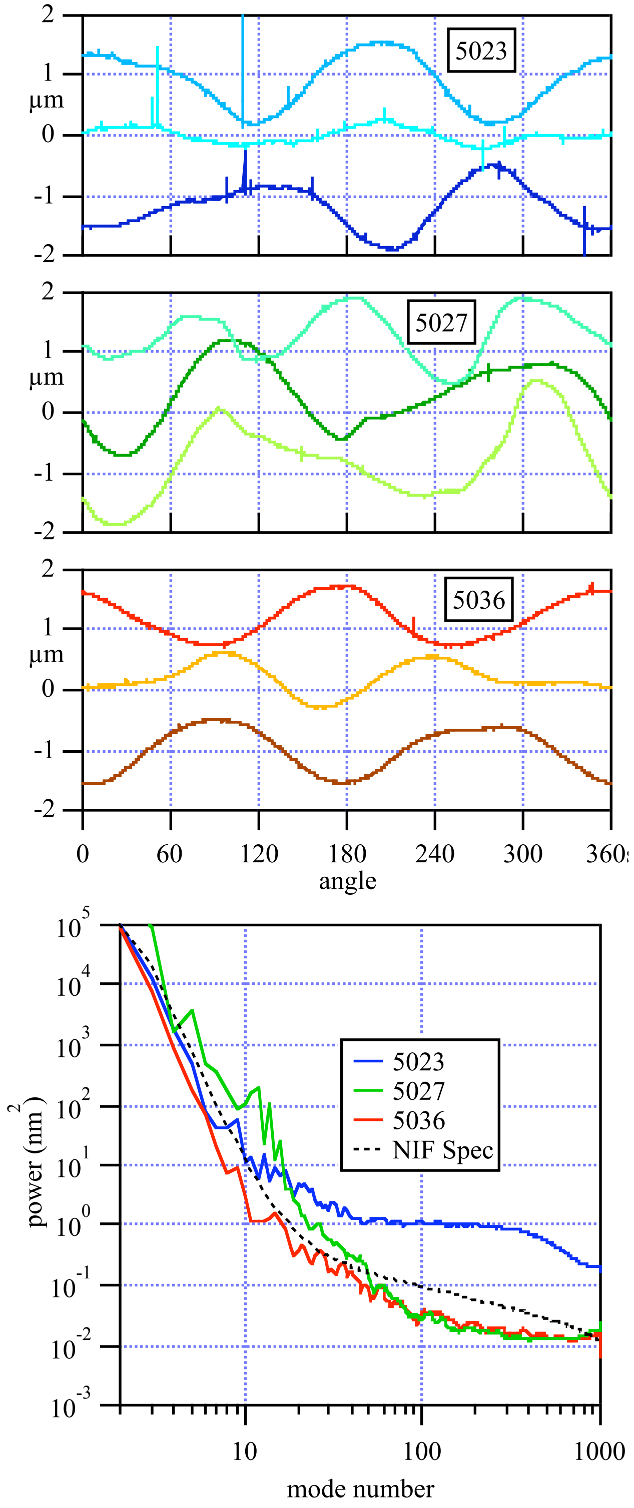

Figure 5. Representative traces and power spectrum from shells prepared by the PAA/PVA combination method and slow cure. The top traces (5023) show problems with surface debris, the middle traces (5027) show problems with mid-mode deformations, and the lower traces (5036) are taken from one of our best 4 mm shells.

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Table I. Summary of 4 mm batches.

Batch Number	g FB in bubbler	Washing method	dry shell OOR (µm)	mode10-100 rms (nm)	mode 101-1000 rms (nm)
5000	60	A'-exchange	6.5	10	10
5001	60	A-exchange	2.1	-	-
5002	60	A-exchange	2.0	8	6
5003	60	A-exchange	2.2	13	21
5004	60	A-exchange	2.4	20	48
5005	60	A"-exchange	2.2	8	6
5006	60	A-exchange	1.0	7	16
5021	60	B-Combination	1.0	10	12
5022	60	B-Combination	1.3	23	5
5023	60	B-Combination	1.8	10	25
5024	60	B-Combination	0.9	10	18
5025	80	C-Combination	1.5	14	8
5026	80	C-Combination	1.4	7	6
5027	80	C-Combination	1.7	15	5
5028	80	C-Combination	1.6	10	11
5029	100	C-Combination	2.6	14	11
5030	100	C-Combination	2.6	6	5
5031	100	C-Combination	3.4	8	13
5032	100	C-Combination	2.8	31	19
5033	120	C-Combination	2.6	37	19
5035	120	C-Combination	2.8	12	28
5036	120	D-Combination	2.5	3	6
5037	120	D-Combination	-	7	3
5038	120	D-Combination	-	26	4
5039	120	D-Combination	-	6	5
5041	100	D-Combination	-	7	16
5042	100	D-Combination	-	33	14
5043	100	D-Combination	-	13	20
5044	100	D-Combination	-	5	7
5045	100	D-Combination	1.8	16	4
5046	100	D-Combination	1.5	12	5
5047	100	D-Combination	1.5	10	5
5048	100	D-Combination	1.5	16	8
5049	100	C-Combination	1.1	5	7
5051	100	C-Combination	1.3	21	6
5052	100	C-Combination	1.3	11	4

Exchange Methods:

A-exchange: Shells are removed from PAA W2 after 72 hours, washed with water, and put into 3% PVA solution to finish the curing

A'-exchange: same as A, but the washing/exchange was at 48 hours

A"-exchange: same as A, but the washing/exchange was at 80 hours

B-combination: at 72 hours, PVA was added to PAA, to have 2 wt% PVA solution

C-combination: at 72 hours, PVA was added to PAA, to have 0.4 wt% PVA solution

D-combination: at 72 hours, PVA was added to PAA, to have 0.2 wt% PVA solution