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HYDRODYNAMIC ISSUES IN PAMS MANDREL TARGET FABRICATION

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Imperfections in PAMS mandrels critically govern the quality of final ICF targets. Imperfections in the mandrels can have a wide range of origins. Here, we present observations of 3 types of imperfections, and data to support the proposal that hydrodynamic factors during the curing of the mandrel are potential causes of these imperfections.

I. INTRODUCTION

The surface finish of a full thickness ICF target depends on the initial symmetry of the PAMS mandrel upon which the ablator layer was coated. Long wavelength surface modulations are reproduced in the final coated shell while short wavelength bump defects generally grow in width during the coating process. The surface finish and symmetry requirements for target quality NIF capsules are exceptionally demanding, thus we are focusing significant effort on perfecting the microencapsulation process that is used to produce the initial PAMS mandrel.

The origins of many of the flaws of PAMS mandrels are puzzling to explain and thereby control. We now understand that the simple picture of solvent leaving the shells by diffusion is a flawed model. Complex fluid dynamics plays a critical role, and can be the origin of defects as well as the source of the high level of symmetry that we observe.

II. MODE 10 BUMPINESS

For several years, the surface of 2mm PAMS mandrels were covered with bumps with an L-mode of about 9–10 (Fig. 1). Figures 1, 3, 6 show three scans of three orthogonal traces around the target. The power in Fig. 1(a) and 2 are shown in 1(b). The origin of these

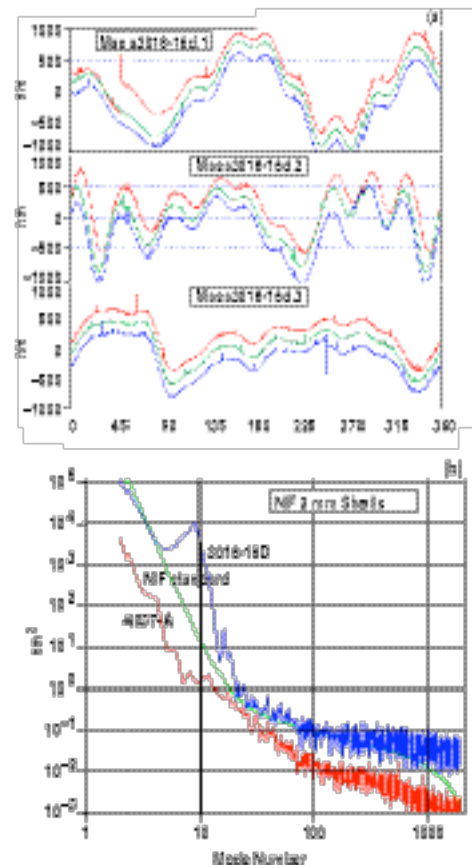


Fig. 1. The scans in 1(a) are shown in the top curve in 1(b).

bumps was puzzling. Ultimately, their origin has been proposed as originating in Marangoni convection.¹ As fluorobenzene leaves the shell wall during curing, convection cells are formed, which pump polymer to the outer surface and form bumps (Fig. 2).

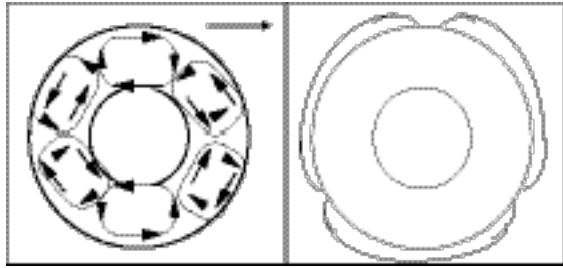


Fig. 2. Representation of convection cells and resulting “mode 3” bumpiness.

The convection cells only form if a combination of parameters exceed a critical value M_c

$$M = (d\sigma/dC)(dC/dL) L^2/\eta D > M_c \tag{1}$$

where $d\sigma/dC$ is the concentration gradient of the outer interfacial surface tension, dC/dL is the radial change of concentration, L is the thickness of the layer, η is the viscosity of the polymer solution, and D is the diffusivity of fluorobenzene in the polymer solution. By adding a bubbler with fluorobenzene, to bubble air with fluorobenzene over the solution of encapsulated shells, we slowed the curing rate and decreased dC/dL , thereby lowering M below M_c . Figure 3 shows how this addition of a bubbler resulted in a much reduced bumpiness across modes 2–20, reduced well below the required NIF standard.

Calculations are underway, to model the Marangoni mode structure throughout the curing process, from first principles. Initial linear theory calculations show that the concentration gradient across the shell wall is very small

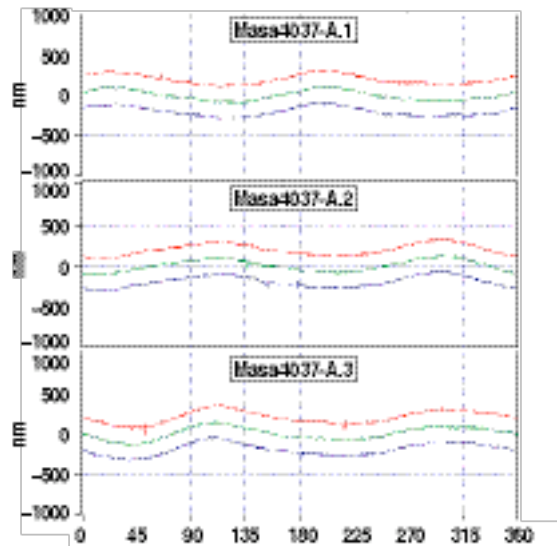


Fig. 3. Shows the shell scans. The resulting power spectrum is shown as the lower curve in Fig. 3(b).

(ΔC of order 0.1% or less of the average concentration), which confirms the lack of formation of an outer skin. The absence of an outer skin allows the outer surface to recede toward the inner interface during curing. In an initial calculation with a linear stability model, where “a single 2 μ m shell” loses fluorobenzene to an infinite bath near saturation ($C_{inf} = 0.0014$, where saturation is 0.0015), with a Biot number of 1.0, the stable modes slowly evolve in time from $L = 1$ to 5 (Fig. 4).

III. PUZZLE WITH CENTERING

Norimatsu has proposed a model for the centering of the inner water drop within the oil drop.² The normal mode oscillation of the compound drop pumps oil (shell wall solution) toward the thin regions, thus pushing the water drop toward the center. We cannot say this model is incorrect, nor can we propose an alternative possibility, but we have observed some data which cannot be explained by his model.

Typically, we add NH_4Cl to the exterior W2 solution (which has PVA), in order to suppress vacuoles and provide density matching in 1 μ m shells.^{3,4} With 2 μ m shells, the suspending W2 has PAA (polyacrylic acid), rather than PVA, and NH_4Cl is incompatible with PAA.⁵ We had sought a salt compatible with PAA, and disodium phosphate is compatible. In seeking to improve 1 μ m shells, we have attempted adding some PAA (and phosphate) to the standard PVA solution. However, the nonconcentricity (NC) of the resulting mandrels has consistently been worse, and out of specifications. Figure 4 shows one example, where six beakers of 1 μ m shells were made (three at the reference 48°C, and 3 at 60°C). We find these results befuddling: why should the addition of salt on the outside of the shell, affect the motion of the interior water ball? We can surmise a change in interfacial tension, but we have not measured the interfacial tension. A decrease in outside interfacial tension may make the

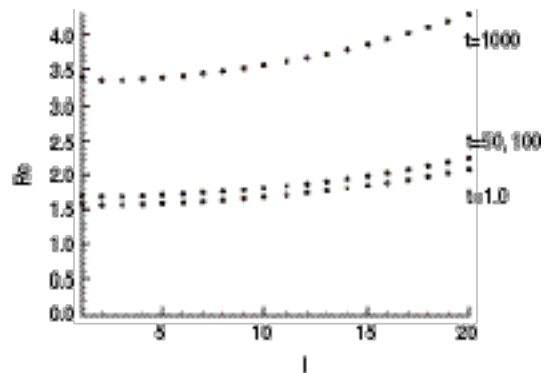


Fig. 4. Marangoni number of each l mode, as a function of time, for a 2 μ m shell curing. Most stable modes are 2, 2, 5 at time = 1, 50, 100, 1000 s.

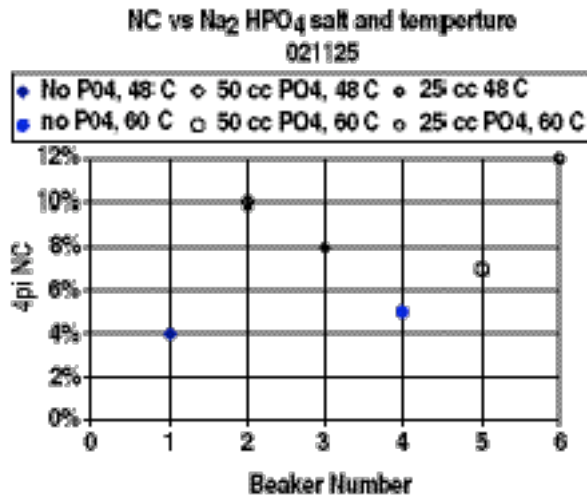


Fig. 5. Beakers 1–3 were at 48°C, 4–6 were at 60°C. Beakers 1 and 4 had no phosphate.

oscillation magnitude larger, thus hastening the centering. So it is possible the interfacial tension is increased upon salt addition, decreasing the oscillation magnitude. Alternatively, the salt may complex an impurity in the oil layer, removing the impurity from the inner interface and changing the inner interfacial tension.

IV. ISOLATED DIP

In some (but not all) spheremapper scans of mandrels, there is a localized singular “dip” (Fig. 6). This dip is typically about 0.5–1.0 μm deep, and 20–40° wide. This dip is actually not a dip in the mandrel surface, but a small change in the radius of curvature over a particular region. This local dip results in a large broad bump in the middle modes of the power spectrum above the NIF specification.

The origin of this bump is unknown. In some mandrels we studied closely, there is one significant vacuole beneath the surface of the dip. Whether this vacuole is the cause of the change in radius of curvature, perhaps by changing the flow pattern near the surface, is unknown. Alternatively, a local change in the interfacial tension would also change the radius of curvature. The cause of such a change in interfacial tension is unknown.

V. SUMMARY

In summary, PAMS mandrels made by micro-encapsulation show several imperfections, whose origins would seem to be in fluid flow effects in the mandrels as they cure. Understanding and modeling these hydrodynamics effects are necessary for making intelligent process changes, for future ICF and IFE targets.

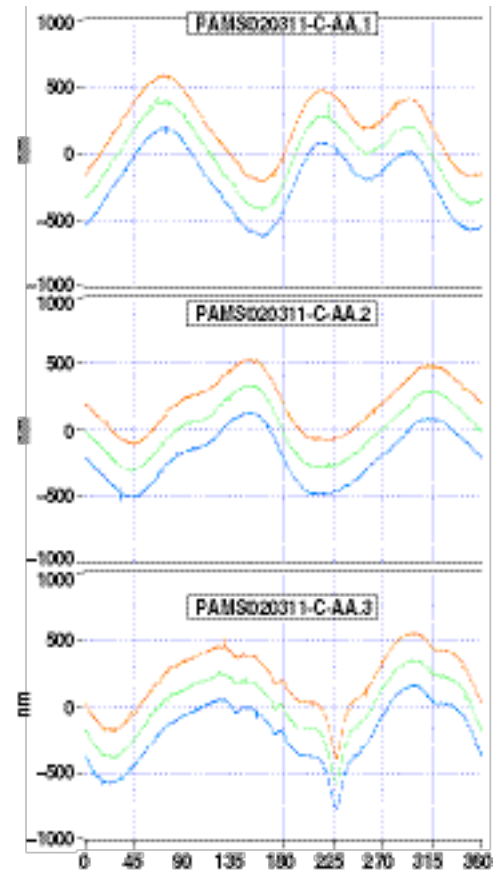


Fig. 6. The isolated dip is seen on the bottom three scans, at about 225 degrees.

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NOMENCLATURE

- PAMS poly(σ-methylstyrene)
- PVA poly(vinyl alcohol)
- C_{inf} The concentration of fluorobenzene in the W2
- Biot Number Bi = k L/D, where k is the coefficient of mass transfer, L is the thickness of the polymer solution, and D is the mass diffusivity of the fluorobenzene in the polymer

W2 the exterior aqueous fluid which suspends the wet shell during its curing. This term is thus distinguished from W1, the water at the center of the shell, and O1 which is the polymer solution which forms the final polymer wall

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