# 1.B   Fabrication of Plastic Shells

Low-atomic-number ablators are a requirement in future direct-drive high-gain capsule designs to reduce preheat; hence, they produce a more efficient compression of the fuel. In the short term, experiments are being planned on OMEGA to investigate the hydrodynamic behavior of capsules with plastic ablators. In this article we will describe the microencapsulation technique now being used to produce the required large numbers of high-quality uniform-thickness plastic shells. The preferred polymer material for production of low-atomic-number capsules is polystyrene due to its chemical and physical stability and its easy processability. Furthermore, it is easily soluble in many common organic solvents at room temperature.

Two methods—both utilizing a polystyrene solution—are commonly used to fabricate polymer shells. In the drop-tower technique,[1] a single-orifice droplet generator is mounted atop a polymer-shell-drying column that has several independently controlled temperature zones. Droplets of polystyrene solution from the generator fall through the column during which time solvent in the outer region of the droplets evaporates and solvent inside is driven off to inflate the shell. The drop-tower method has not yet, however, been able to fabricate polymer shells with wall-thickness uniformity comparable to that of glass shells.

Microencapsulation, the second method, was developed by U. Kubo[2] of Kinki University, Japan in 1981. A schematic of the process is illustrated in Fig. 42.8. Polystyrene solution and water are rapidly agitated in a small closed vial to produce an emulsion of water droplets suspended in the polystyrene solution. This emulsion is then poured into heated and agitated water containing a polyvinyl alcohol (PVA) surfactant into which the emulsion dispenses forming droplets of water surrounded by a thin spherical layer of polystyrene solution. The temperature is then raised to accelerate the evaporation of solvent from the polystyrene. This results in solid polystyrene shells that contain pure water. These shells are then rinsed in pure water to remove residual PVA surfactant and air dried to remove internal water.

Microencapsulation has been extensively refined at Osaka University's Institute of Laser Engineering (ILE) to the extent that they are currently capable of producing target-quality polymer shells.[3] ILE now fabricates not only polystyrene shells but also deuterated polystyrene and polytrimethyl silylstyrene shells.[3] Using ILE's procedure as a general guideline, LLE has developed a process that yields target-quality polystyrene and deuterated polystyrene shells in large quantity. While ILE's and LLE's methods of fabrication are essentially the same during initial stages of processing, they differ in the method of water removal from the shell interior. In the ILE process, shells are rinsed with pure water and air dried in a filter funnel until all interior water has permeated through the shell wall. While this process does produce uniform shells, many are cracked, perhaps due to water permeating through the shell wall too rapidly. The process results in the

- W/O emulsion

Water globule

Polystyrene
solution

- W/O/W emulsion

Polyvinyl alcohol
in H_2O
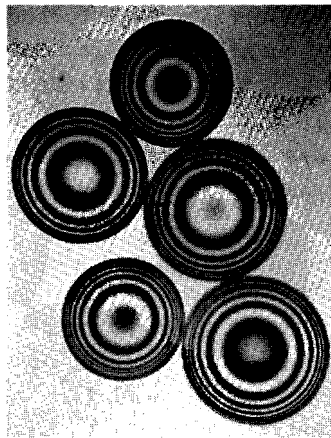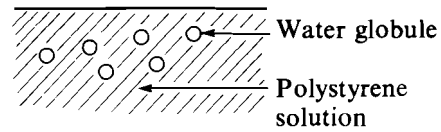
Polystyrene
solution
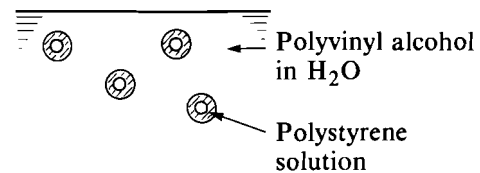
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Fig. 42.8
Schematic of microencapsulation technique of forming plastic shells. W/O (water in oil) emulsion is first formed in which water globules are dispersed in polystyrene solution. This is followed by a W/O/W emulsion in which shells of polystyrene solution containing water are dispersed in polyvinyl alcohol solution in water.

time-consuming operation of optically inspecting each shell for cracking. In the LLE process, shells are rinsed in pure water to remove residual PVA and then placed in a dilute mixture of ethanol and water. This allows for accurate rate control of water removal from the interior of the shells via osmotic action. This can be accomplished because styrene is highly permeable to water, while being impermeable to ethanol. Although some shells still crack during water removal, they are automatically removed from the batch because they sink. Only shells that do not crack or have very minor cracks float to the surface where they are removed by pipette and placed in a grid to complete drying in a vacuum oven.

In practice, polystyrene solutions are made using polystyrene fractions having a narrow molecular-weight distribution, either alone or in combination with other molecular-weight fractions. These molecular-weight fractions are typically in the 100,000–600,000 range. Similarly, the solutions are made in a variety of weight percentages ranging from 3 to 7 wt.%. The PVA solution is made using 25,000-molecular-weight polymer at 5 wt.% concentration. A 50/50 volume ratio of benzene and ethylenedichloride is the solvent used for polystyrene. Both the polystyrene and PVA solutions are filtered to 0.2 μm to reduce contamination in the shell wall and on its surface. Approximately 8 cc of filtered polystyrene solution is prepared in

a 15-cc vial. This is capped and heated to ~50°C for a period of 2 h to allow the polymer to degas. After cooling to room temperature, 4 cc of filtered water is placed in the vial. The mixture is recapped and shaken vigorously by hand for 10 s. The resulting emulsion is poured rapidly into the heated PVA solution that is agitated by a downward pushing propeller-type stirrer rotating at ~400 rpm. After ~10 s, the propeller speed is reduced to 200 rpm. At this time the PVA bath temperature is raised from its initial temperature of 72°C to a final temperature of ~85°C. After ~3 h at the elevated temperature, all of the benzene and dichloroethane is removed and the shells are rinsed, sieved, and placed in the alcohol/water solution for interior-water removal.

Plastic shells are target quality if they meet the three following conditions: (1) the shells are spherical and the wall is of uniform thickness; (2) the rms surface roughness is less than 1 μm; and (3) the density of the shell wall is uniform, i.e., no vacuoles are present in the shell wall. Generally, a single batch contains up to several thousand shells and 10% to 50% of them are "target quality" shells. Figure 42.9 shows Mäch-Zehnder interferograms of polystyrene shells of different sizes (diameters of 216 μm, 265 μm, 373 μm, and 640 μm). These shells are from a single batch and in any given batch, the shell size ranges, in general, from 100 to 700 μm. The wall thickness

OD:
216 μm

WT:
3.1 μm

OD:
265 μm

WT:
7.3 μm

OD:
373 μm

WT:
4.4 μm

OD:
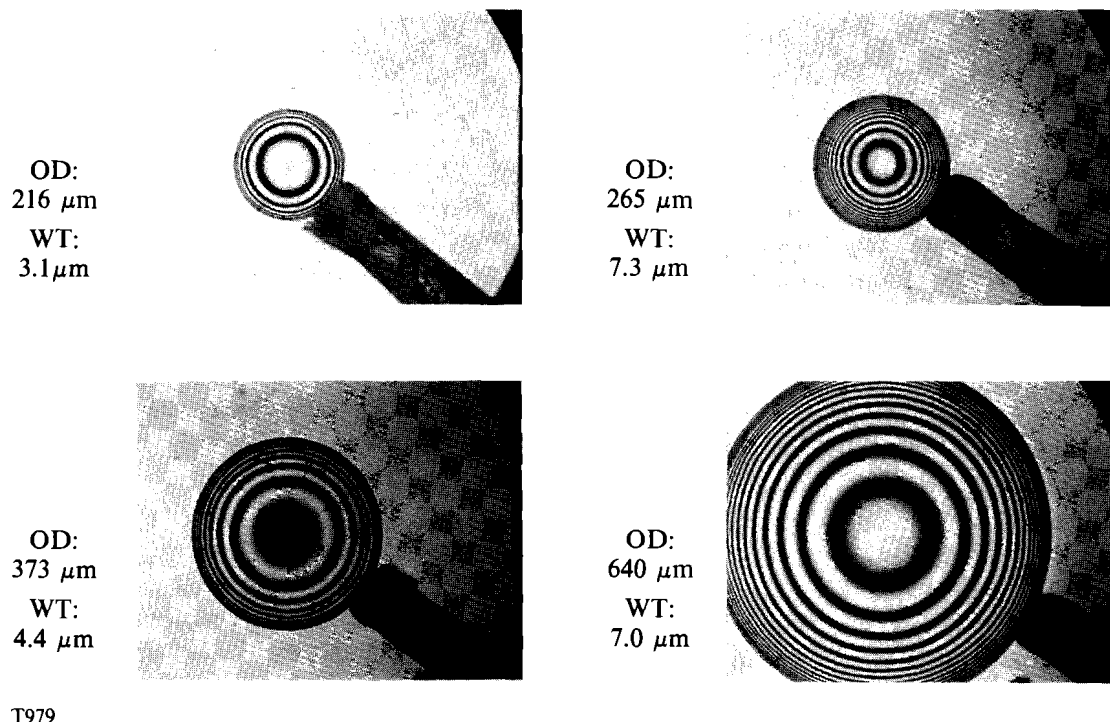640 μm

WT:
7.0 μm

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Fig. 42.9
A typical batch of polystyrene shells with a wide range of sizes and wall thicknesses.

ranges from 2 to 7 μm. Production of sizes and wall thicknesses of the shells outside the above-described ranges may require further study. Concentricity of the shells was analyzed by comparing the interferograms from two orthogonal views with templates generated by a ray-tracing computation. Figure 42.10 shows a Mäch-Zehnder interferogram, a template, and their superposition. From this comparison, this particular shell is shown to have a nonconcentricity of 2%, which is well within the acceptable range of 5%. Figure 42.11 is a scanning electron microscope (SEM) photo of a shell showing the surface finish. The object in the middle of the photo is believed to be a dust particle deposited on the surface. The surface is very smooth with an rms finish much smaller than 1 μm. Although not all shells have such a smooth surface finish, a sufficient number of shells of good surface quality can be selected from any given batch.

LLE plastic shell   Template   Superposition
OD: 216 μm
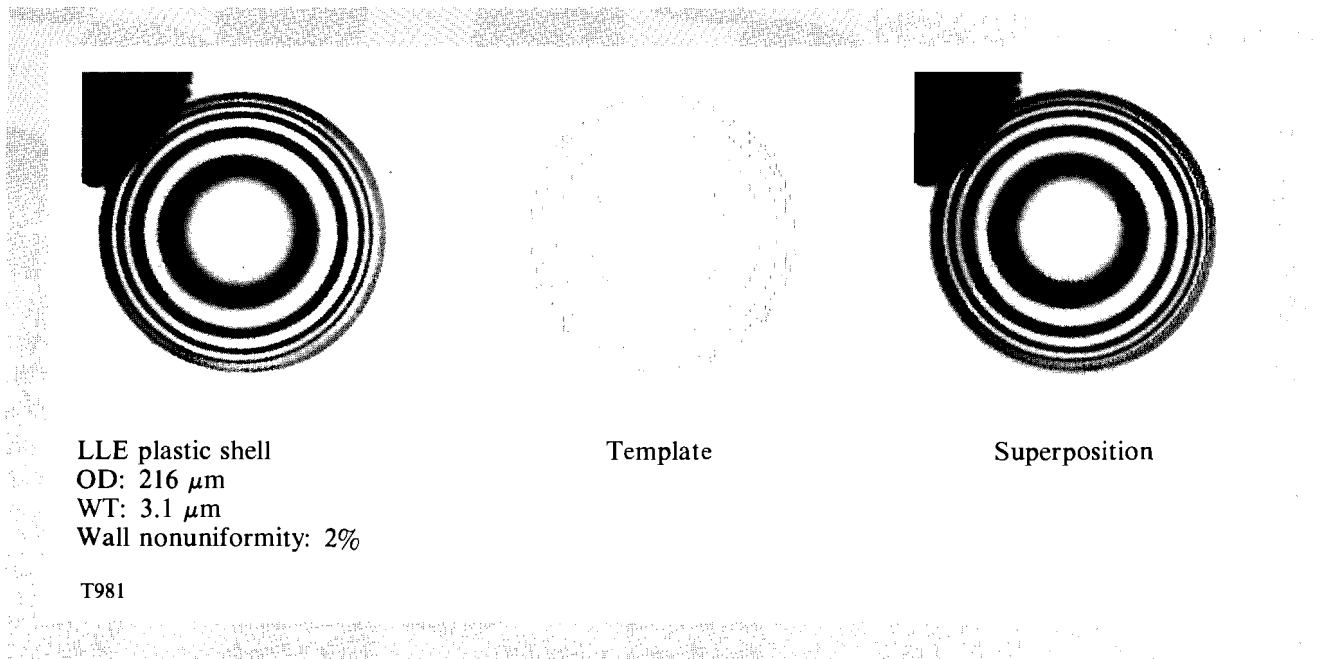WT: 3.1 μm
Wall nonuniformity: 2%

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Fig. 42.10
Characterization of a polystyrene shell. Interferogram is superposed onto a template calculated from a ray-tracing program. From this comparison, the shell is shown to have a nonconcentricity of 2%.

Nonuniformity of plastic shell density is generally caused by the presence of vacuoles in the wall that can be incorporated during the fabrication process. In fact, problems with vacuoles caused the abandonment of some early attempts to develop the microencapsulation technique.[4] However, in our experiments, it was found that the number of vacuoles can be reduced or eliminated by simply degassing the polymer and reducing the size of the container holding the mixture of water and polymer solution. Optical microscopy is utilized to detect the vacuoles in the shell. When a shell is illuminated with plane-parallel wave fronts, the transmitted rays either converge or diverge depending upon whether the defect is surface debris or vacuoles embedded in the shell, respectively. Therefore, by focusing
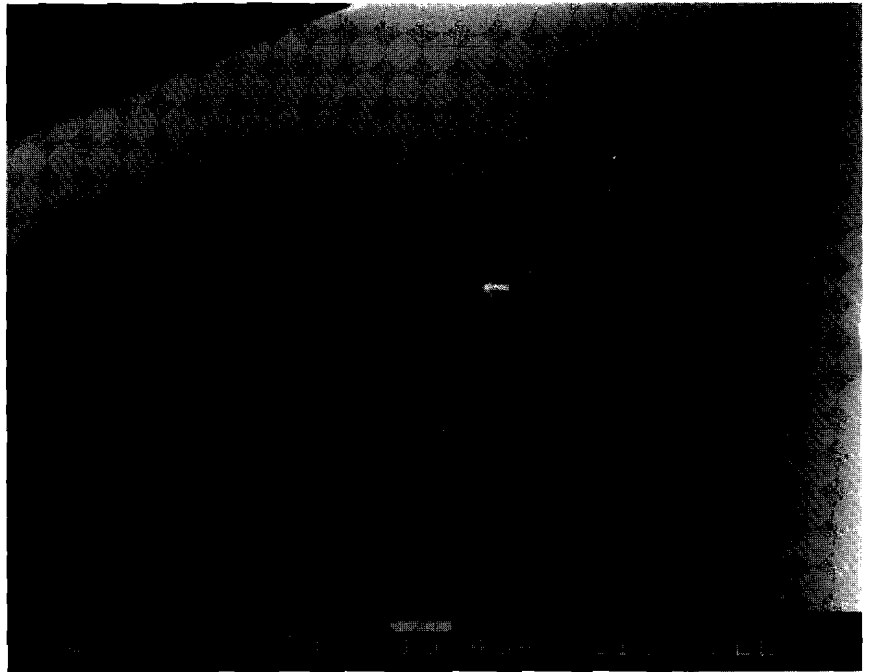
T980

Fig. 42.11
Surface finish of a polystyrene shell. The defects in the surface are artifacts.

through the object plane and noting whether the bright point of light appears above or below the plane in which the object is clearly in focus, one can easily differentiate between surface debris or vacuoles. However, because of the refractive effects of the shell wall itself, this technique is useful only for regions of the shell on which the light from the condenser is normally incident. The shell must therefore be rotated to examine it entirely. Typical polymer shells produced at LLE contain only a few vacuoles, if any, which are of the order of up to several microns in diameter.

In summary, using the microencapsulation technique, it is possible to fabricate a large number of polystyrene shells. The fabricated shells have very uniform wall thickness, smooth surface finish, and few vacuoles embedded in the wall. The size and wall thickness of the shells range from 100 to 700 μm and from 2 to 7 μm, respectively. We have also succeeded in fabricating deuterated polystyrene shells. Since deuterated and protonated polystyrenes are chemically identical, the same procedure also produced target-quality deuterated polystyrene shells. This present work will be expanded to fabricate the large-size shells needed for OMEGA Upgrade experiments and plastic shells containing Si, Cl, or other elements.

REFERENCES

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4. KMS Fusion Annual Technical Report, KMSF-U-1728 (1985), p. 73.