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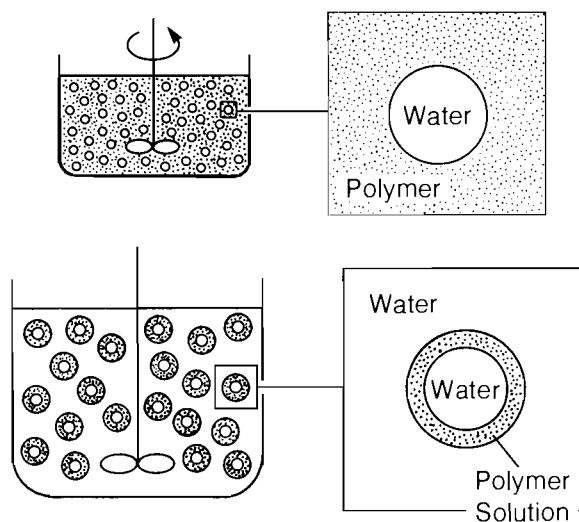
2.B Polymer Microballoon Development

For many years inertial-confinement-fusion (ICF) experiments have been conducted with DT-filled, glass microballoon targets. The attractive properties of glass shells are their low permeability to DT gas and relatively high tensile strength, which enables them to withstand fairly high internal pressures. However, the use of high-Z silica glass promotes radiative preheat of the fuel when illuminated by a UV laser. This reduces the maximum obtainable fuel compression. Low-Z polymer microballoons can minimize this effect and also provide uniform ablative surfaces. Moreover, experimental diagnosis of the fuel core is easier because low-Z polymers are more transparent to the soft x rays generated during the implosion. But for ICF target fabrication, the polymer microballoons must have mechanical properties comparable to those of glass, i.e., high tensile strength and low permeability.

Two methods of polymer shell fabrication are currently being investigated at major ICF laboratories. First, there is the drop or spray tower method in which a liquid-phase bubble is generated at the top of a vertical column and passed through a heated zone to drive off the solvent. The solid polymer shell is collected at the bottom of the tower in a water bath. The second technique is to microencapsulate water with a polymer solution and then drive off the solvent to produce a shell. This second method is the one we are pursuing here at LLE.

This particular microencapsulation technique was developed by Kubo¹ and is depicted in Fig. 20.10. The polymer is solvated in a low-boiling-point, high-vapor-pressure solvent that is normally immiscible or slightly soluble in water. Under continuous agitation a water phase containing approximately 50 ppm of surfactant is emulsified in the polymer solution. The resulting emulsion is a continuous polymer (oil)

phase with water droplets uniformly distributed throughout. After 20 min of agitation with a mechanical stirrer the droplet size is relatively uniform, with an average diameter that is a function of the speed of agitation. The emulsion is then poured into a second water phase containing approximately 600 ppm of surfactant. The result is a suspension in the second water phase of small polymer "bubbles" containing the first water phase. This solution is heated to drive off the polymer solvent, leaving polymer shells containing the first water phase. Finally, the shells are collected by sieving and gently heated to remove the interior water.



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Fig. 20.10
The microencapsulation process for producing polymer-shell microballoons.

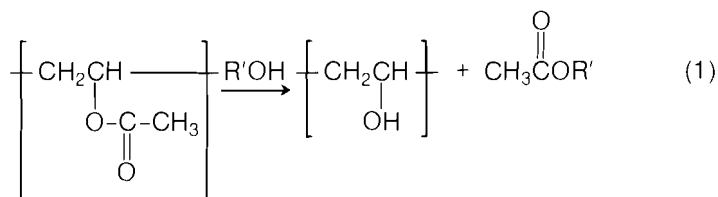
The focus of the University of Rochester's LLE microencapsulation program is on materials development rather than process development. Originally, Kubo's work was conducted with polystyrene (PS), a polymer which is easily solvated and exhibits favorable characteristics for microencapsulation. However, PS is a poor target material because of its relatively high permeability [23.3×10^{-10} cc hydrogen $\text{cm}/(\text{cm}^2 \cdot \text{sec} \cdot \text{cm Hg})$] and low tensile strength (40–70 N/mm²).² More suitable polymers include polyacrylonitrile (PAN), polyvinyl alcohol (PVA), and regenerated cellulose, all of which exhibit permeabilities that are five orders of magnitude lower than that of PS and tensile strengths that are ten times greater. To date, most of the polymer microballoon development work at LLE has focused on PAN because it is relatively easy to process and is highly resistant to water. PVA and cellulose are known to be hydrophilic and quickly deteriorate upon exposure to water.

Good solvents for PAN generally have high boiling points and low vapor pressure, except for nitromethane which is similar to water. However, solvating PAN in nitromethane is difficult, requiring the application of heat (greater than 80°C) and continuous agitation for three to

four hours. Even after this period not all the polymer is in solution, and micron-sized particles which affect the quality of the balloons are present. To resolve this, a PAN blend of 94% PAN and 6% methyl acrylate (provided by E. I. du Pont de Nemours & Co.) has been used. This reduces the processing temperature to 60°C and the blend is more easily solvated than pure PAN. The anticipated increase in permeability due to the use of the copolymer has not yet been determined but it is expected to be less than one order of magnitude—an acceptable increment.

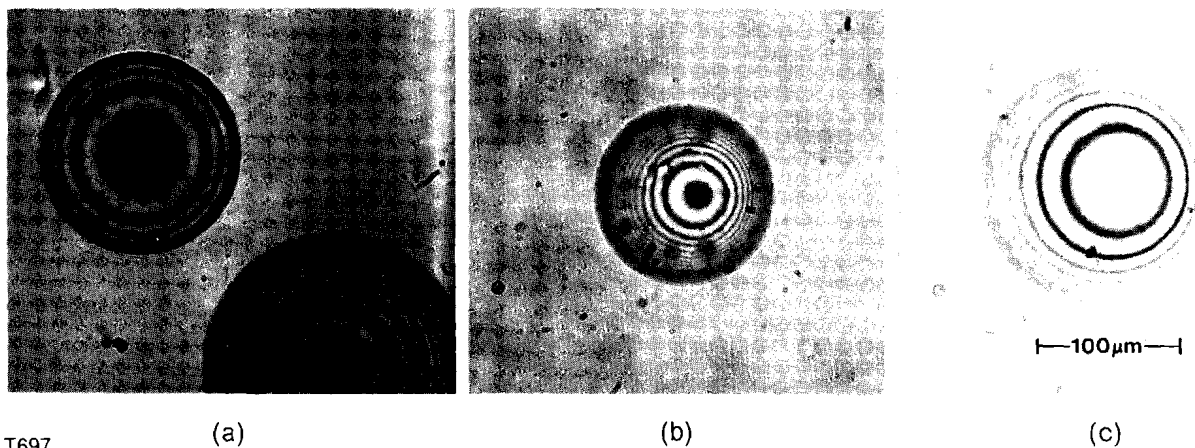
We have performed experiments with a solvent system consisting of 85% nitromethane, 10% water, and 5% 2,2,2-trifluoroethanol. The polymer is pulverized in a laboratory blender to a 20- μm size and introduced to the solvent at room temperature. The solution is then heated to 70°C and slowly agitated for one to two hours in a closed system. The first water phase is heated to the same process temperature before addition, and the speed of agitation is increased to achieve the desired shell size. After the required agitation time, the emulsion is siphoned through a heated line into the second water phase which has also been heated to 70°C. Care must be taken to keep the process temperature and the temperature of all equipment coming in contact with the solution above 60°C, or the solution begins to gel and particles form. The pressure may be reduced somewhat, depending on the temperature of the system, to accelerate solvent removal. When most of the solvent has been driven off, the micro-balloons are collected and gently dried at 35–40°C for 48 to 72 hours.

Polyvinyl acetate (PVAc), although it has better physical properties than PS (tensile strength = 30–50 and permeability = 8.9×10^{-10}) is still not suitable for target material. However, PVAc is easily converted to PVA through a transesterification shown in Eq. 1. The conversion of PVAc to PVA has not yet been seriously addressed at LLE but will be in the future.



PVAc, like PS, is easily solvated by most solvents so that the process conditions are similar. The one notable exception is that the final suspension is cooled to 15–20°C to prevent coagulation as a result of heat softening of the polymer. (The softening point of PVAc is 28–31°C.)³

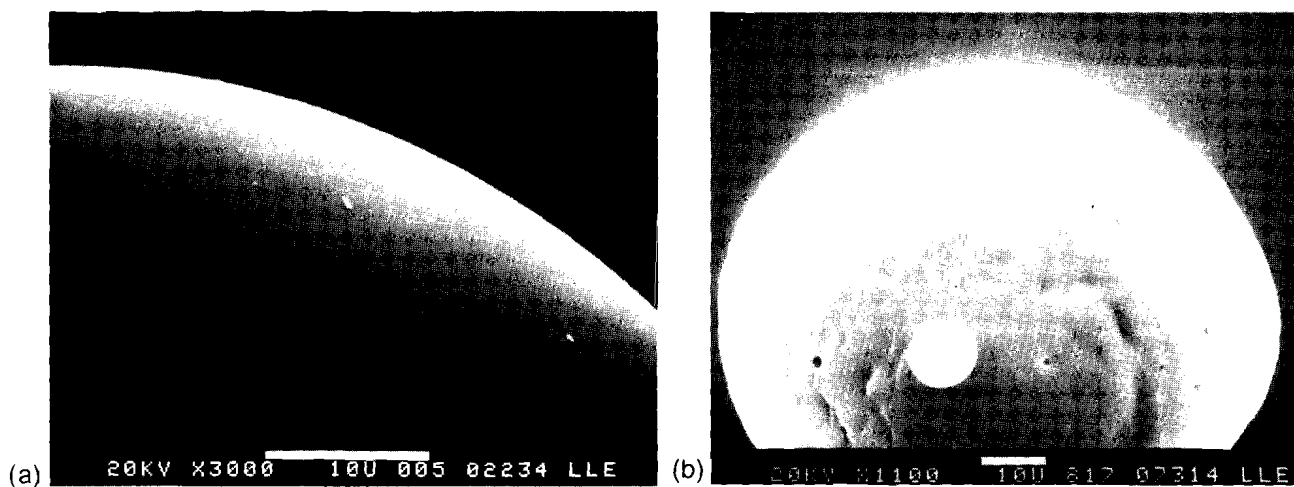
In experiments with PS, PAN, and PVAc, we have successfully produced target-quality balloons 50–400 μm in diameter, with wall thicknesses of 5–25 μm . Non-concentricity and sphericity are typically less than 5% (Fig. 20.11). The degree of success varies with the material. PS has been by far the most successful, with more than 50% yield of target-quality balloons. PAN, on the other hand, yielded only



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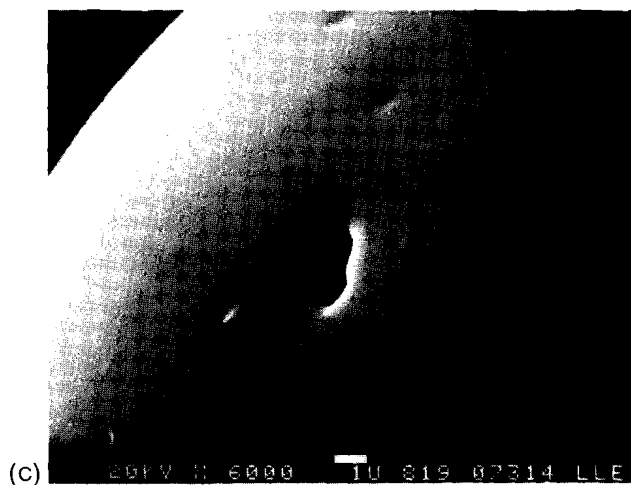
Fig. 20.11
Interferograms of polymer microballoons
manufactured using microencapsulation:
(a) polystyrene
(b) polyacrylonitrile copolymer
(c) polyvinyl acetate

10-20%; this is attributable to the lack of complete polymer solution. With respect to surface finish, both PS and PVAc microballoons exhibit surface roughnesses of less than 250 Å, but the presence of micron-sized particles degrades the surface finish of PAN microballoons (see Fig. 20.12).



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Fig. 20.12
Scanning-electron micrographs of poly-
mer microballoons manufactured using
microencapsulation:
(a) polystyrene
(b) polyacrylonitrile
(c) polyacrylonitrile copolymer



Gentle removal of the interior water phase is very important if the microballoons are to survive. Overly vigorous drying tends to cause stress in the balloons and produces small surface cracks like those depicted in Fig. 20.13. This effect has been observed in microballoons with wall thicknesses less than 10 μm for removal temperatures greater than 50°C.

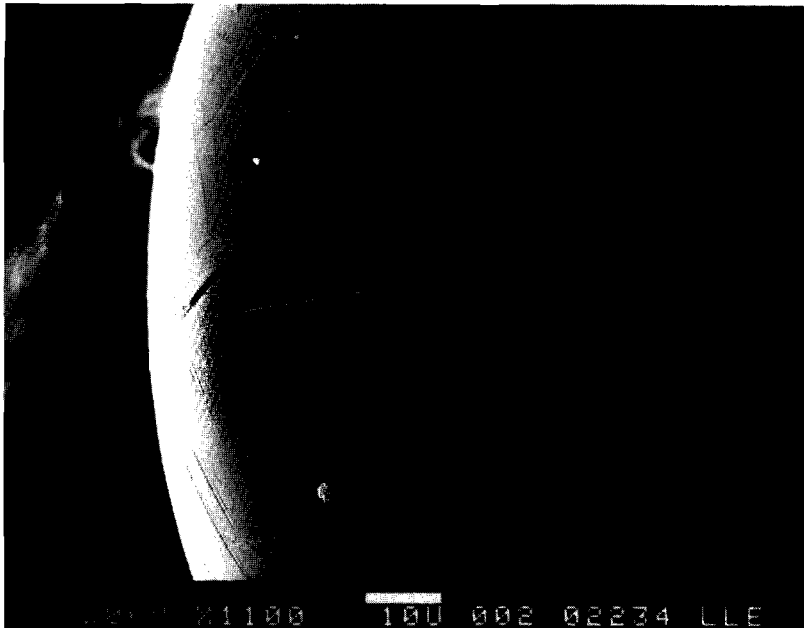


Fig. 20.13
Scanning-electron micrograph of the surface of a polystyrene microballoon from which the interior water was removed too violently.

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In short, we have employed microencapsulation techniques to fabricate target-quality microballoons with the polymers PS, PAN, and PVAc. To date, the highest yields and best surface finishes have been obtained with PS and PVAc. Although PAN microballoons are more difficult to produce, we continue to focus on this material because it is stronger and less permeable to hydrogen gas. We also plan to work further to explore the hydrolysis of PVAc to PVA.

ACKNOWLEDGMENT

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