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Polymers Replace Glass in Nova Fuel Capsules

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Polymers Replace Glass in Nova Fuel Capsules

The relatively small fuel capsules used in past laser-fusion research were made of glass, a material that will not be suitable for the advanced implosion-physics experiments to be done on Nova. We are developing large polymer capsules to meet the new physics requirements.

The glass fuel-capsule designs used in our previous laser-fusion research are not adaptable to the implosion-physics requirements of Nova and other more powerful laser facilities that may be available in the future. Historically, glass capsules have been used because they are a convenient way to contain the deuterium-tritium (D-T) commonly used as fuel. However, glass capsules do not work well because of hydrodynamic instabilities and inefficiency that results from radiative heat loss associated with high-Z material. Moreover, the glass impedes passage of x rays used to diagnose the D-T plasma. As we try to learn more about the physics of high-density compression, it becomes increasingly important to replace the glass with lower-Z material. Accordingly, we have shut down the high-temperature drop-tower furnaces we used to make glass capsules, and we are focusing all our efforts on developing new techniques for making polymer capsules. These capsules are ten times larger in diameter than the glass capsules used in the early days of laser-fusion research, but they are still only one-tenth as large as a high-gain capsule must be.

The polymer capsules will be used in classified "indirect-drive" targets. Although unclassified laser-fusion experiments, both at the Laboratory and elsewhere, drive the fuel capsule by focusing a powerful laser beam or beams directly on it, we believe that such a direct-drive approach cannot illuminate the capsule uniformly enough to achieve the high compression needed for high gain. Instead, we have concentrated our experimental implosion program on the indirect-drive approach, in which the laser light is shone not on the capsule but on the inside of a high-Z-metal radiation case, or hohlraum, containing the capsule. In the hohlraum, the light is converted to a uniform blackbody-like radiation field of soft x rays, which are absorbed by the outer wall of the fuel capsule, causing it to ablate and thus imploding the capsule and the D-T fuel contained inside.

These polymer capsules will be used first as gas-filled targets to refine our understanding of the implosion

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physics, then later in a cryogenic version where the liquid or solid D-T fuel coats the inside of the capsule in a uniform layer. The layered target is hydrodynamically equivalent to a high-gain target.[1] The leading candidate for a high-gain target is actually a hydrocarbon foam shell that acts as a wick to hold liquid D-T. Such cryogenic foam targets are not currently planned for use in Nova, however. Instead they are being evaluated in experiments carried out in underground nuclear tests, in the Halite program.

This article describes how the decisions were made on which polymers to use in our Nova fuel capsules, the techniques explored, and the properties of the prototype capsules.

Design Issues for Polymer Capsules

In choosing polymers for Nova implosion capsules we considered implosion characteristics, diagnostic compatibility, physical properties relating to fielding, and ease of fabrication.

Hatchett and Lindl[2] recently detailed the design issues for the indirect-drive implosion capsules for Nova. The simplest capsule design they studied, a thick capsule of hydrocarbon (CH_x) polymer, was among the best performers. Other good designs had a higher opacity layer within the capsule wall to serve as a preheat shield.

The most obvious implosion diagnostic needed is one that can measure the number of fusion neutrons generated, which is proportional to target gain. However, to determine which of the many possible mechanisms are degrading target performance, it is also desirable to have methods of determining other parameters such as the density, ρr, and symmetry of the fuel as well as the dynamics, symmetry, and stability of the fuel/pusher interface. Our laser experiments group has proposed a battery of neutron and x-ray diagnostics to measure these parameters. The diagnostics require D-T in the fuel capsule, to produce 14-MeV neutrons, and a low-opacity capsule wall to allow x rays to escape. Finally, some of the diagnostic methods require that inner parts of the capsule be doped with bromine or boron or that the fuel be doped with a high-Z gas, such as krypton.

Polymer capsules can meet all these diagnostics requirements if properly chosen. However, some polymers are susceptible to D-T radiation damage, some are excessively permeable to the fill gas they must contain (ironically, the least permeable polymers are among the more sensitive to radiation), and some have insufficient strength to withstand the pressure of the fill gas. Therefore we must choose our polymers carefully and characterize their physical properties thoroughly.

A final issue is the availability of the capsules. They should either be obtainable from commercial suppliers or be reasonably easy to fabricate at the Laboratory. Both glass and polymer microballoons are available commercially, but essentially none of these meet target specifications. Substantial resources were spent at KMS Fusion and LLNL in refining the technique to fabricate glass shells for laser targets, so high-quality glass shells are available over a wide range of specifications. Less extensive work on polymer shells has demonstrated that they can be made from a wide variety of polymers by either droplet-drying or microencapsulation. Polystyrene is one of the easiest materials to work with in terms of availability, ease of dissolution, and yield of high-quality shells. Polyacrylonitrile, polyvinyl alcohol (PVA), and carboxymethylcellulose (CMC) have been explored extensively because of their low permeability, but high-quality shells have been produced only from PVA, and these only in small sizes, in limited quantities, and with considerable difficulty.

About two years ago, we considered all these factors and

decided to pursue an all-CH capsule using an inner polystyrene shell as thick as we could make with sufficient wall uniformity. There were two main reasons. First, fabricating large plastic shells of the very high quality required for Nova appeared to be a difficult task, and information in the literature as well as our own experience indicated that polystyrene shells would be the easiest to make. Second, we had insufficient data available to make a quantitative assessment of the radiation damage effects on polymer capsules, but we knew that a polystyrene-based capsule had the highest chance of survival because of its short fill time (due to high permeability) and its high radiation tolerance. In the short term, a simple cryogenic system would enable us to study implosions of capsules filled with deuterium (D_2) gas, and cryogenic transport looked like a feasible way of delivering the D-T-filled target capsule to the target chamber. In case we were unsuccessful, KMS Fusion would continue to investigate the fabrication of Nova-sized shells from alternative polymers and glass.

As a result of this decision, we spent a year and a half developing the techniques to fabricate these capsules and measuring the physical properties necessary to fill them with fuel and minimize its leakage. Most of this article describes the results of that work. However, one of the problems with this approach is that the cryogenic systems are very costly to implement. We therefore began developing an alternative capsule design that will enable us to study gas-filled implosions without the need for cryogenics and that will also be hydrodynamically equivalent to a reactor-scale target capsule. This alternative design involves coating the polystyrene shell with a thin, impermeable layer of PVA before adding a thick layer of plasma-generated CH coating for the ablator. The polystyrene inner shell serves as a barrier to the undesirable implosion characteristics caused by having the highest wall density and opacity next to the D-T fuel. The balance of this article discusses our initial attempts to fabricate this new capsule design.

Fabrication of Polymer Implosion Capsules

With the initial decision to use an all-CH-polymer fuel capsule for Nova, we began a substantial effort to learn how to fabricate such a target. Since it was doubtful that the capsule could be made to the proper thickness and concentricity in a single step, our strategy was to develop methods of making thinner polystyrene shells, which would then be coated with plasma-generated CH polymer to the final dimensions. Since coating is a very slow process (1 μm/hr), we wanted to produce shells that were as thick as possible. Because the polystyrene and CH coating compositions and densities are fairly close, and because it has been possible in the past to eliminate concentricity defects by the coating procedure, it appeared that shells with highly spherical centers might be adequate and that the highly uniform wall thickness required for glass shells might not be necessary. However, wall smoothness would still be required to prevent coating defects.

Making the Polystyrene Shells

Polystyrene shells have been made by several processes. We evaluated two techniques in detail: droplet-drying and microencapsulation. Droplet-drying is essentially the same process we used for many years to make glass shells. Microencapsulation is a process for fabricating tiny capsules of one material to contain another. It has many other applications[3] besides the making of hollow polymer laser targets.[4–6] We evaluated these two processes in parallel. When we began our investigations, we expected that the microencapsulation process would have a better chance of producing polystyrene shells nearer to the desired thickness, shells that would require less coating to bring them to final thickness.

We started our microencapsulation work using a double-emulsion technique that had been used to produce polystyrene shells with diameters less than 500 μm. We quickly found, however, that this method produced shells too small for Nova. Therefore we modified the method by designing a dual-orifice droplet generator similar to one we had used previously to make PVA shells in a drop tower. In this approach, concentric jets of polymer solution and water are formed, which break up within a centimeter into water droplets surrounded by polystyrene solution. The solvent dissolves into the water, and the hardened capsules are dried in a vacuum oven to remove the water. We made polystyrene shells from 300 to 1000 μm in diameter, with wall thicknesses up to 50 μm, but we were never able to achieve the necessary quality control. Therefore, we eventually abandoned this microencapsulation approach in favor of droplet-drying.

In the droplet-drying method, polystyrene shells are made by drying droplets of polystyrene solution as they fall through a heated column.[6,7] Our studies indicate that this is the best technique for producing high-quality polystyrene shells of Nova dimensions.[1] The apparatus we used (Fig. 1) has a column 15 cm in diameter and 4.5 m long, with 11 heated zones that can be controlled at temperatures up to 300°C. We recently upgraded the column so that the bottom section has a refrigerant recirculation system capable of maintaining −70°C. There are inlets for refrigerated gas at the bottom and preheated gas at the interface between the cooled and heated sections. The upper portion is heated typically to 200°C at atmospheric pressure with a slow upward flow of purge gas (usually nitrogen at 4 L/min).

We selected methylene chloride as the primary solvent for three reasons: it has a relatively low boiling point of

40°C, it is an excellent solvent for polystyrene, and it is not flammable.

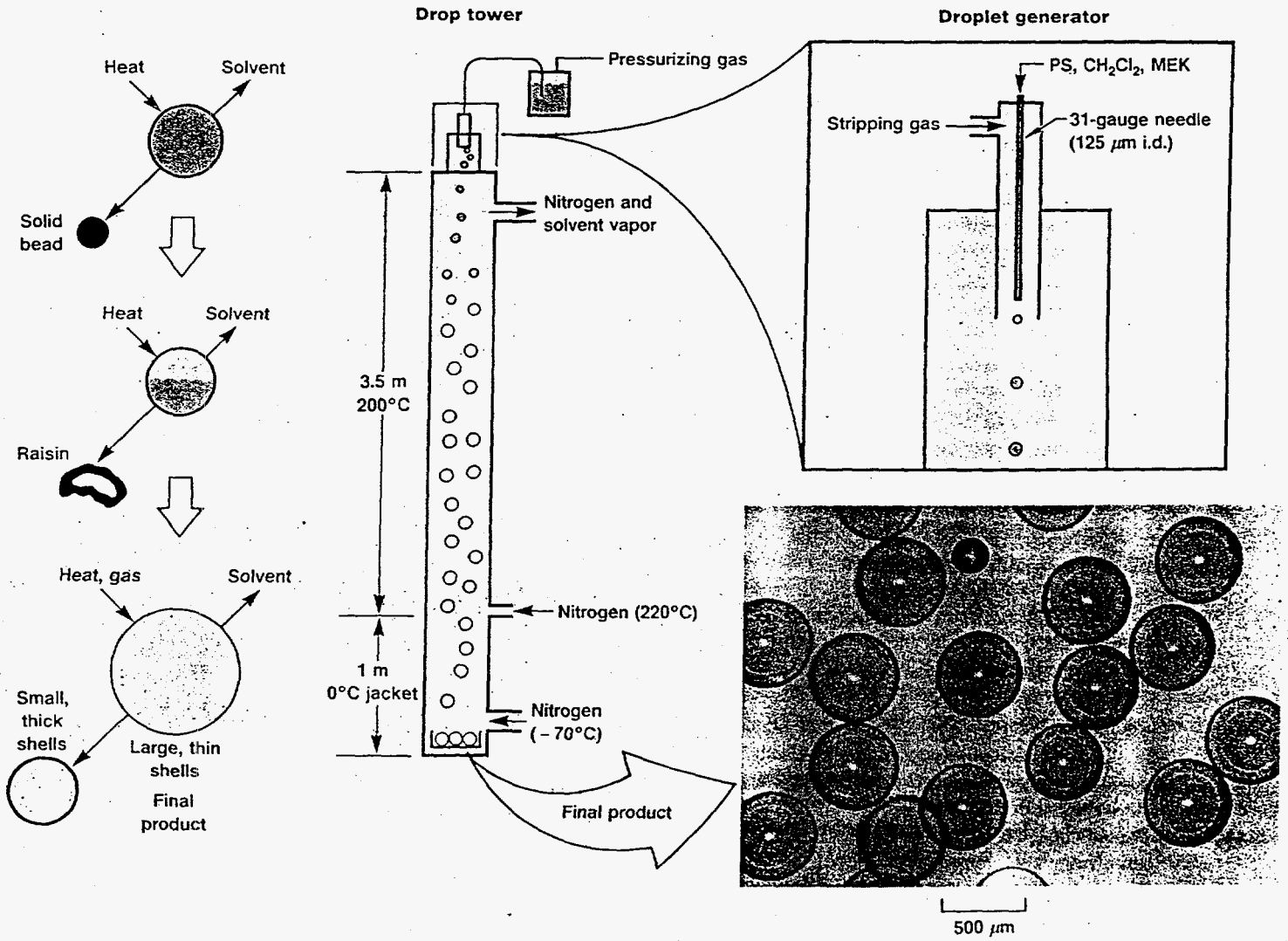
Devising a reliable generator to feed droplets of polymer solution into the column proved difficult. The polymer solutions have complicated flow properties and are quite viscous above 10% polystyrene. Since methylene chloride does not readily charge, our initial efforts using the traditional design of an ultrasonic generator with electrostatic charge and deflection were unsuccessful. However, we were successful when we used a stream of solvent-saturated air to strip droplets from a capillary tip. These droplets were sufficiently spaced so that there was no problem with collisions in the column. Moreover, we were able to make droplets in the desired size range with variation of less than 10%. While this degree of uniformity is lower than that obtainable with ultrasonically driven jet breakup, it is adequate for our purposes.

Our first attempts at making Nova-size polystyrene shells produced more defective products than good ones. The unwanted products were of four types: raisins, solid beads, doughboys (dented shells), and small, thick shells. Initially, we developed a simple procedure for concentrating the high-quality shells. Static electricity makes sieving impossible, so we developed a vacuum microsweeper to quickly separate the products of interest.

Three improvements led to the increased yield of high-quality shells.

Fig. 1

Apparatus for producing polymer microshells by the droplet-drying process. Conditions leading to the various final products, which include both desired and undesired products, are illustrated schematically. PS is polystyrene, MEK is methyl ethyl ketone.



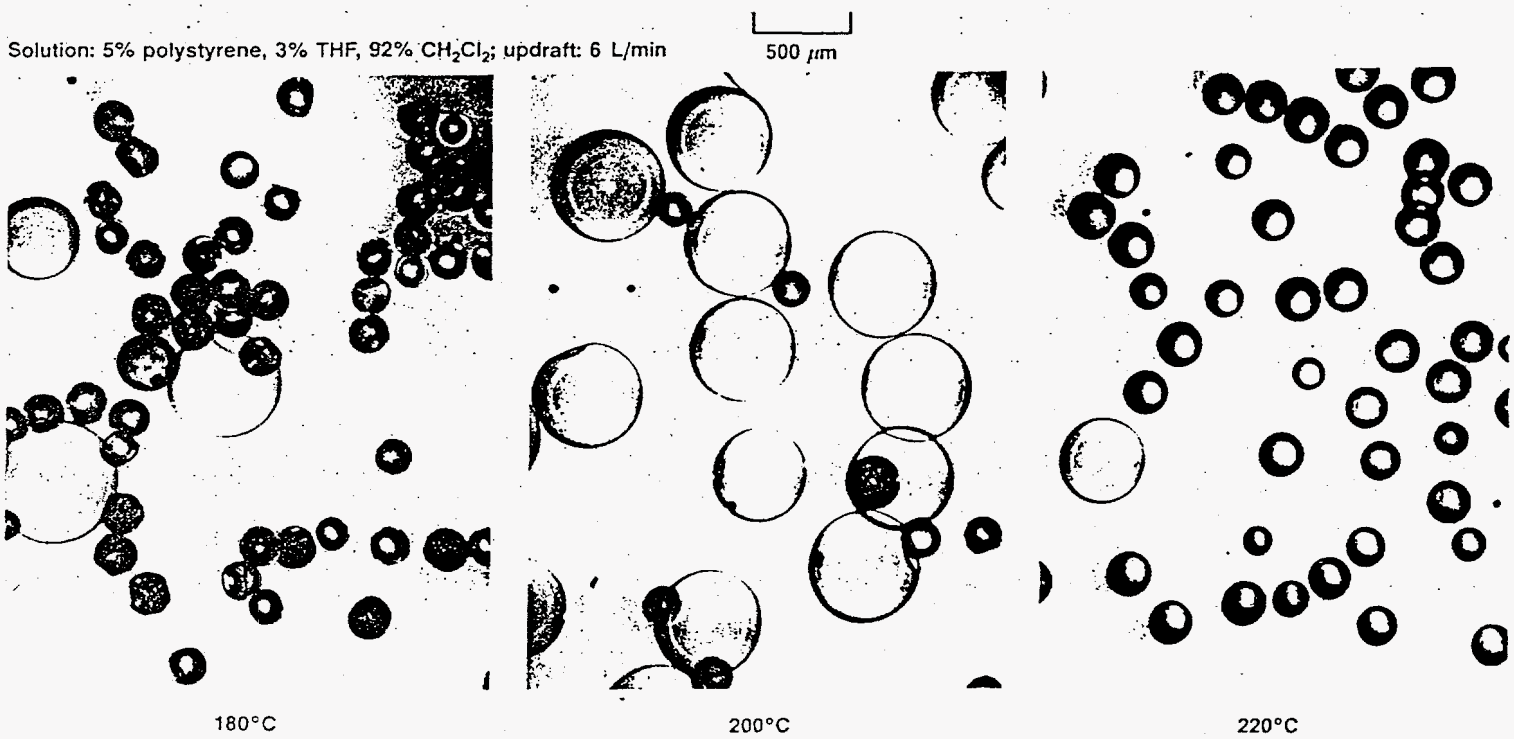
We first noticed that the fraction of solid beads increased during the course of a run, and we assumed that a buildup of solvent vapor was inhibiting skin formation on the drying droplet, thereby preventing the shell-blowing process. Improvements in the gas inlet and exit systems largely eliminated the solid beads. We learned how to minimize the raisins and small thick shells by studying the effects of drying temperature in detail. The product distribution of three runs is shown in Fig. 2. The raisins are obviously due to too low a temperature, and the small, thick shells are apparently caused by shrinkage of the large, thin shells when they are heated too much. We were unable to find conditions that would increase the small, thick-walled shells to Nova size, so we concentrated on improving the yield of the large, thin shells. Fine-tuning the column temperature largely eliminated the raisins and small, thick shells.

The doughboy defect is an inward local buckling of the shell. We thought it was caused by slow cooling. The gas cooling inside the shell contracts, and the polystyrene wall is too warm and soft to maintain a pressure difference, so it buckles inward. We solved this problem by controlling the cooling in the column. We cooled the bottom section of the column to −70°C, causing the shell wall to cool faster than the interior gas. This allows the wall to increase its strength more before it must support a significant pressure drop. We can now make much larger shells without doughboy defects.

We have examined several batches of shells in detail. Scanning electron microscopy indicates that the surface finish is generally better than 50 nm, although occasional defects 20-fold larger are present. The sorted batches typically have diameter variations of ±5% and wall thickness variations of ±10%. The batch-average concentricity defect determined from single-axis interferometry is typically about 20%, which corresponds to less than 1 μm for the thin shells. Multi-axis interferometry of a batch of 430-μm-diameter shells with 4.4-μm-thick walls revealed that about half of them had maximum concentricity and ellipticity defects less than 1 μm and 0.25 μm, respectively. If these shells could be coated smoothly and

Fig. 2

Effect of drying temperature is shown in these samples of shells produced in three runs at different temperatures. THF is tetrahydrofuran.



uniformly, they would probably be satisfactory as targets.

We also developed a fabrication technique that doped the polystyrene shells with bromine. Bromine has been proposed as a replacement for silicon in glass shells, in order to permit the use of neutron activation to measure shell ρr. We first attempted to add the bromine by exposing the polystyrene shells to bromine vapor. However, the bromine softened and distorted the shells before substantial exchange with hydrogen occurred. We next tried adding small amounts of liquid bromine to the polystyrene solution before processing. With this technique, we found that adequate bromine (a few tenths of a percent) could be incorporated into the polystyrene without affecting the shell-forming process.

Coating the Shells

The CH ablator is added to the polystyrene shells by plasma polymerization. A mixture of trans-2-butene and hydrogen is fed into a glow-discharge plasma, where a variety of free radicals are formed, and a highly cross-linked CH polymer is deposited on the shells below.[8] To ensure a uniform coating, the shells are subjected to random movement during this deposition by being bounced in a pan vibrated by a piezoelectric crystal that is driven by a white-noise voltage source. If the amplitude of the vibration is tuned properly, it is even possible to reduce shell concentricity defects because the thicker side tends to spend a longer time on the bottom and the coating rate is higher on the top.[9]

Polystyrene shells made by both the microencapsulation and drop-tower techniques were coated using this technique. Shells produced by microencapsulation and the small, thick shells from the drop tower were easier to coat than the large, thin shells from the drop tower. The latter had a tendency to agglomerate during the coating process, producing very nonuniform coatings. This behavior is apparently due to the high ratio of static charge to mass for the large, thin shells. Unfortunately, we were not able to either reduce the vacuoles in the microencapsulation shells to an acceptable level or increase the diameter of the small, thick shells from the drop tower to Nova size, so we attempted to circumvent the static electricity problem of the drop-tower shells.

Increasing the vibration amplitude was unsatisfactory because the amplitude needed to keep the shells separated was greater than that required to bounce them out of the pan. Increasing the wall height of the pan permitted higher amplitudes but drastically slowed the coating rate. We had better success by reducing the static charge, which we accomplished by pretreating the shells with an antistatic agent and starting the coating run with a period of hydrogen plasma stripping. Sufficient quantities of coated shells were produced by this technique for mechanical testing and quality testing. A photograph of one batch coated to a wall thickness of 80 μm is shown in Fig. 3. The efficiency of this coating procedure was lower than desired, but improvements were not pursued because of a programmatic shift to the capsule containing the PVA interlayer for a permeation barrier.

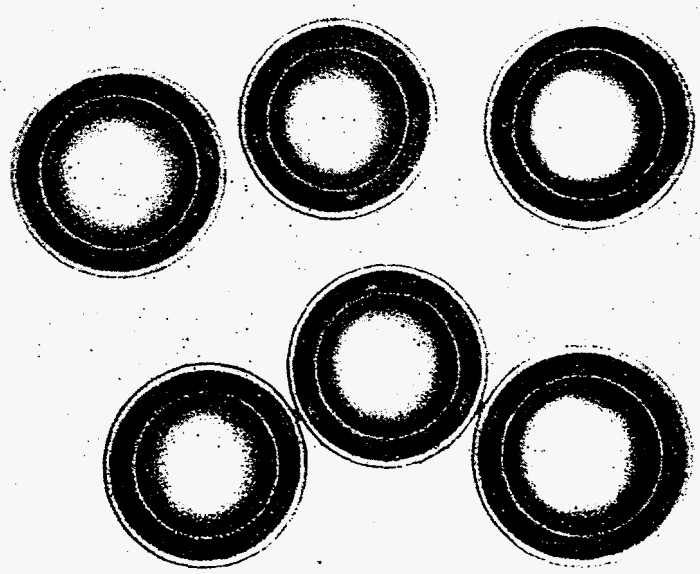


Fig. 3

Samples from a batch of shells coated by the plasma polymerization method to a wall thickness of 80 μm, near that required for a target capsule.

The permeability of PVA to hydrogen is 1000 times less than that of polystyrene. Annamalai et al.[10] report attempts to coat polystyrene shells with PVA by exposing a shell levitated in a focused acoustic field to an aerosol of aqueous PVA. However, this technique is complicated, and it resulted in poor-quality coatings. Therefore we explored alternative approaches, all involving embedding a polystyrene shell in a drop of PVA solution, then drying it in a drop tower at mild enough conditions so that the PVA film would not blister during the drying process. The best approach is shown in Fig. 4. The polystyrene shells are sorted for quality and vacuumed into a capillary tube. Then the PVA solution is drawn into the capillary with a syringe. The loaded capillary is inserted into a gas nozzle so that a coaxial flow of gas strips the solution-coated shells from the capillary tip as they are pumped out. A column temperature of 100°C produced uniform, blister-free coatings with a surface finish better than 0.1 μm. Single-axis interferometry indicated that many of the coated shells had a concentricity defect of less than 1 μm on a 500-μm-o.d. shell. The thickness of the coating can be changed by varying the ratio of the shell and capillary diameters or the solution concentration. We have not determined how thick we can make coatings before gravity causes unacceptably large coating nonuniformity. In principle, thin alternating coatings of PVA and polystyrene could be applied to achieve sufficient total PVA thickness to reduce permeation to an acceptable level. Moreover, it appears likely that other polymer coatings containing elements for diagnostic purposes could be applied at various stages, with the only restriction being that they must be soluble in a solvent that does not damage the last shell layer. A bonus of the PVA coating is that it drastically reduces the static electricity problem and makes the shells far easier to coat with the plasma-generated CH polymer.

Filling the Capsules

The only practical way developed so far to fill the tiny target capsules with gas is by the diffusion-loading technique. In this process, the empty capsules are put into a pressure chamber, which is then pressurized with the gas that is to be loaded into them (usually D-T). The gas diffuses through the walls of the capsules, from the outside in, and fills them to the desired internal pressure. Heat may be applied during this process to increase the permeability of the capsule walls to the gas.

Several factors are important in determining the response of the capsules to being filled with gas. The permeabilities and compressive and tensile strengths of the inner shell and its coating—all of which are temperature-dependent—determine the fill time and temperature, the maximum fill pressure, and the temperature required to maintain the fill gas in the capsule. We have measured the room-temperature

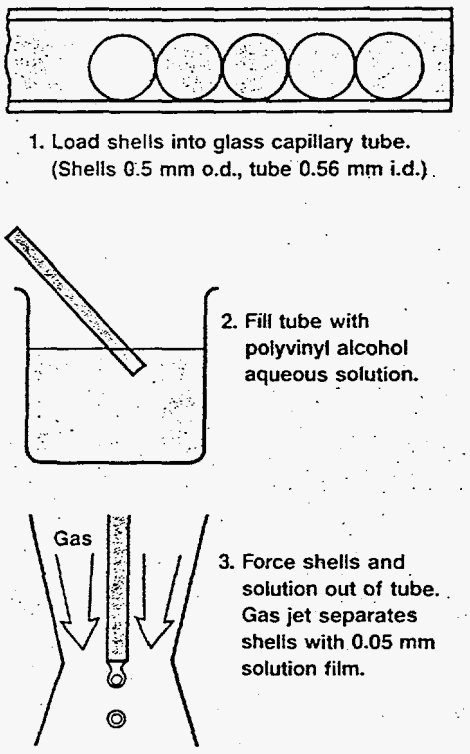


Fig. 4

Process for coating polystyrene shells with polyvinyl alcohol (PVA), which is 1000 times less permeable to hydrogen.

permeability to hydrogen, helium, and argon, the room-temperature compressive and tensile strengths, and the tensile strength at liquid-nitrogen temperature for uncoated and coated polystyrene shells. Combined with literature data and previous measurements at LLNL on PVA shells, these results provide an excellent basis for evaluating and comparing the procedures required for fielding all-CH and PVA-interlayer polymer implosion capsules.

Governing Equations

The time required to fill an implosion capsule depends on the capsule's permeability to the gas of interest and its compressive strength. The fill rate is proportional to the pressure drop across the capsule wall, which for a constant external pressure decays with a time constant τ given by

$$\tau = \frac{(r_o - r_i)r_i^2}{3RTKr_o}, \qquad (1)$$

where r_i and r_o are the inside and outside radii, K is the permeability, R is the gas constant, and T is the absolute temperature.

If the capsule is strong enough to support a pressure drop higher than the desired fill pressure, it can be filled with a single pressure step by allowing sufficient time (about five time constants) for the internal and external pressures to approach equilibrium.

The maximum external pressure that a capsule can withstand depends on the ratio of the capsule's wall thickness to its diameter. Thin shells collapse by elastic buckling, and the ideal failure pressure is given by

$$P_f = \frac{8E\left(\frac{r_o - r_i}{2r}\right)^2}{[3(1 - \nu^2)]^{1/2}}, \qquad (2)$$

where E is Young's modulus and ν is Poisson's ratio. This represents an upper bound for real situations, and stretching, cracks, or complete failure can occur if the strain in the buckling region exceeds the elastic limit. A lower bound, normally called the minimum buckling pressure, can be estimated from the empirical equation

$$P_f = 1.46E\left(\frac{r_o - r_i}{2r}\right)^{1/2} \qquad (3)$$

As the shell becomes thicker, there is a transition to an inelastic-shearing failure mode. This failure pressure is given by

$$P_f = 2S_c\left(\frac{r_o^3 - r_i^3}{r_o^3 + 2r_i^3}\right) \simeq 4S_c\left(\frac{r_o - r_i}{2r}\right) \qquad (4)$$

where S_c is the compressive strength.

If the desired fill pressure exceeds the compressive failure pressure of the capsule divided by a safety factor of four or so, the capsule can be filled in a series of steps, or, even better, by ramping the external pressure so that a constant pressure drop is maintained across the wall. The minimum time required for a ramp fill is equal to $\tau P/\Delta P$, where P is the desired fill pressure and ΔP is the constant pressure drop. Due to variations in the permeability of different shells, however, it is wise to hold them at the fill pressure for two or more time constants to increase fill accuracy. On the other hand, if the fill pressure is much lower than the failure pressure, as in the case of a trace diagnostic, and some variation in the fill pressure is allowable, the fill time can be significantly shorter than the time constant.

When the external pressure on the capsule is released after the capsule is filled, an excessive amount of the fill gas must not leak out before the target is shot and the capsule must not crack or burst due to tensile failure. The rate of leakage is characterized by the same time constant given in Eq. (1). Since the permeability of materials increases exponentially with temperature, this implies generally that the capsules must be stored at a substantially lower temperature than that at which they are filled. It is therefore necessary to

know the temperature dependence (activation energy) of the permeability to choose the appropriate fill and storage temperatures. The tensile failure pressure of the capsule is also given by Eq. (4), except that the compressive strength is replaced by the tensile strength, S_t. Materials generally become stronger as temperature decreases, so the temperature dependence of the capsule strength should also be known.

Capsule Tests

We developed a simple apparatus to determine the filling and leak characteristics of the CH-coated polystyrene capsules. We suspected that the permeabilities of the polystyrene and the CH coating were close enough that neither would dominate the effective capsule permeability, so both would have to be determined. Our objective was to have a system that could measure the leak rate of a single capsule or a few capsules at a time, because we could only produce them in small quantities and because we thought we might need to determine the permeability of each capsule to be used for an implosion experiment to ensure proper filling. The resulting apparatus is shown in Fig. 5. Basically, a microtransducer measures the pressure

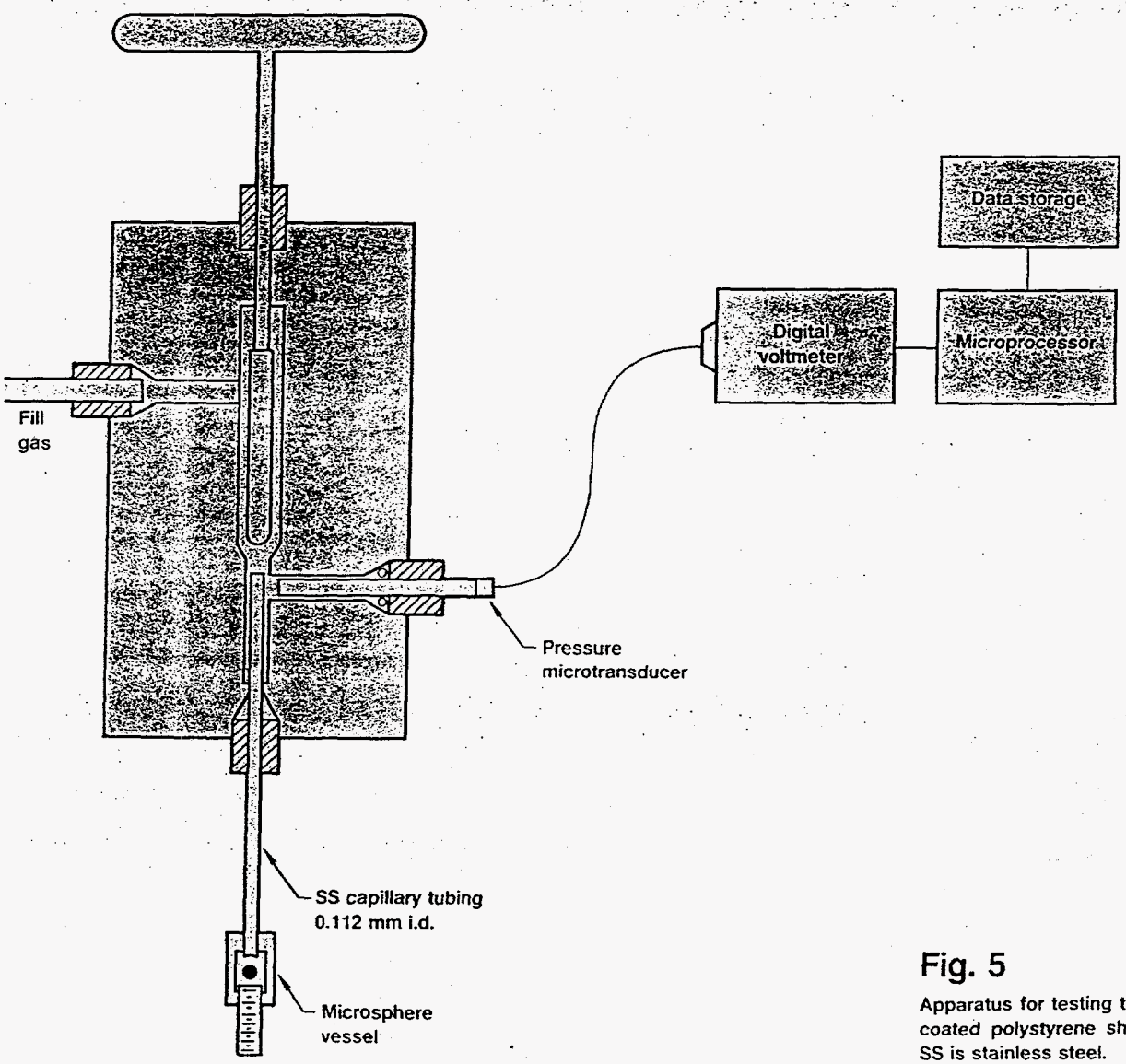


Fig. 5

Apparatus for testing the permeability of CH-coated polystyrene shells to various gases. SS is stainless steel.

rise in a microvolume due to leakage of the fill gas. Preliminary results of these tests at room temperature are given in Table 1 along with data from handbooks ("literature data") and other sources.[11] This is the first demonstration that the CH coating is more permeable than polystyrene.

The apparatus used for compressive strength tests is shown in Fig. 6. The cell pressure and the video image of several capsules glued to a glass slide are displayed on a monitor and recorded on a video cassette recorder as the cell is pressurized. This must be done quickly so that a negligible quantity of gas permeates into the capsule during the pressurizing step. The exact compressive failure pressure of each capsule is determined by reviewing the video tape.

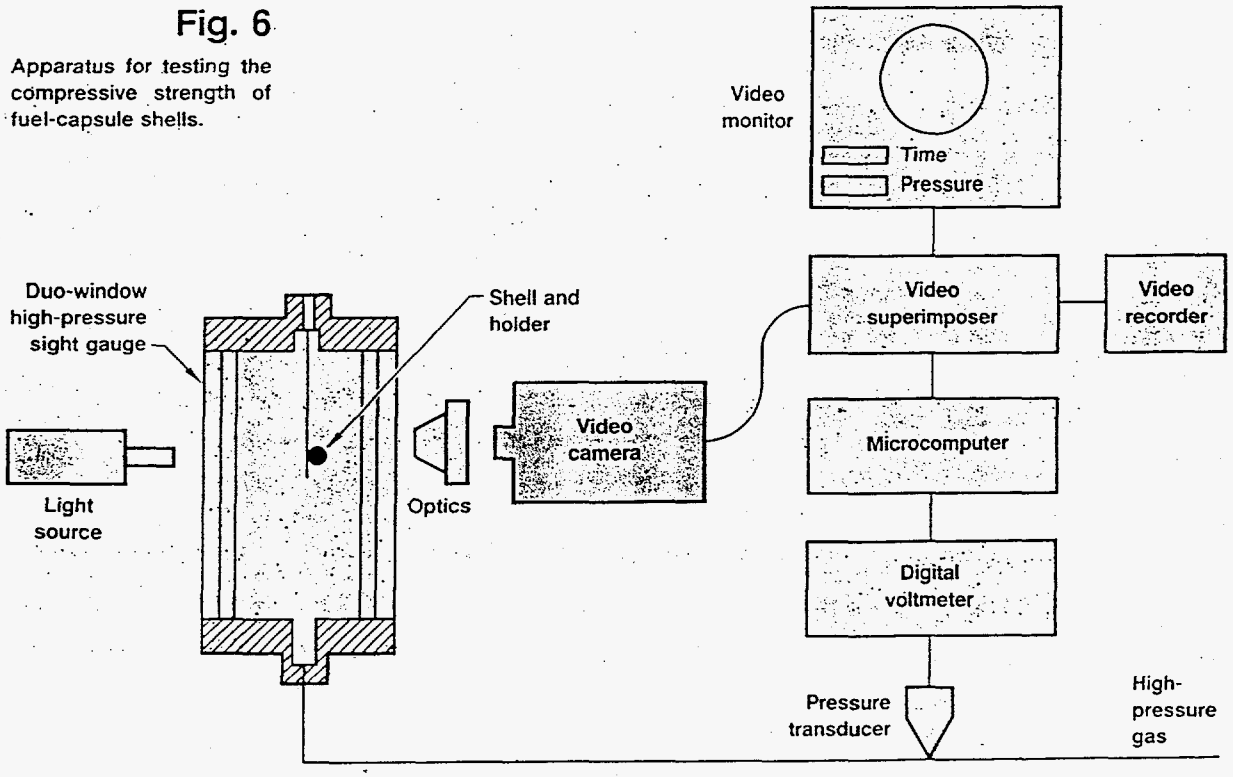
The results of these tests are summarized in Fig. 7. The outside diameters ranged from 250 to 560 μm

Table 1 Permeability of polystyrene shells and CH coating to various gases, in units of 10^{-15} mol·m/m^2·s·Pa.

Source	Polystyrene shell			CH coating	
	Argon	Helium	Hydrogen	Helium	Hydrogen[b]
Our measurements[a]	0.35	2.1	2.7	11	7
KMSF[a,b]	—	6.2	6.9	—	—
Isakov[a,11]	0.08	—	4.0	—	—
Literature[c]	0.25	5.0	4–7	—	—

[a]Measurements on shells.
[b]Personal communication from L. Scott of KMS Fusion, Inc.
[c]Measurements on films.

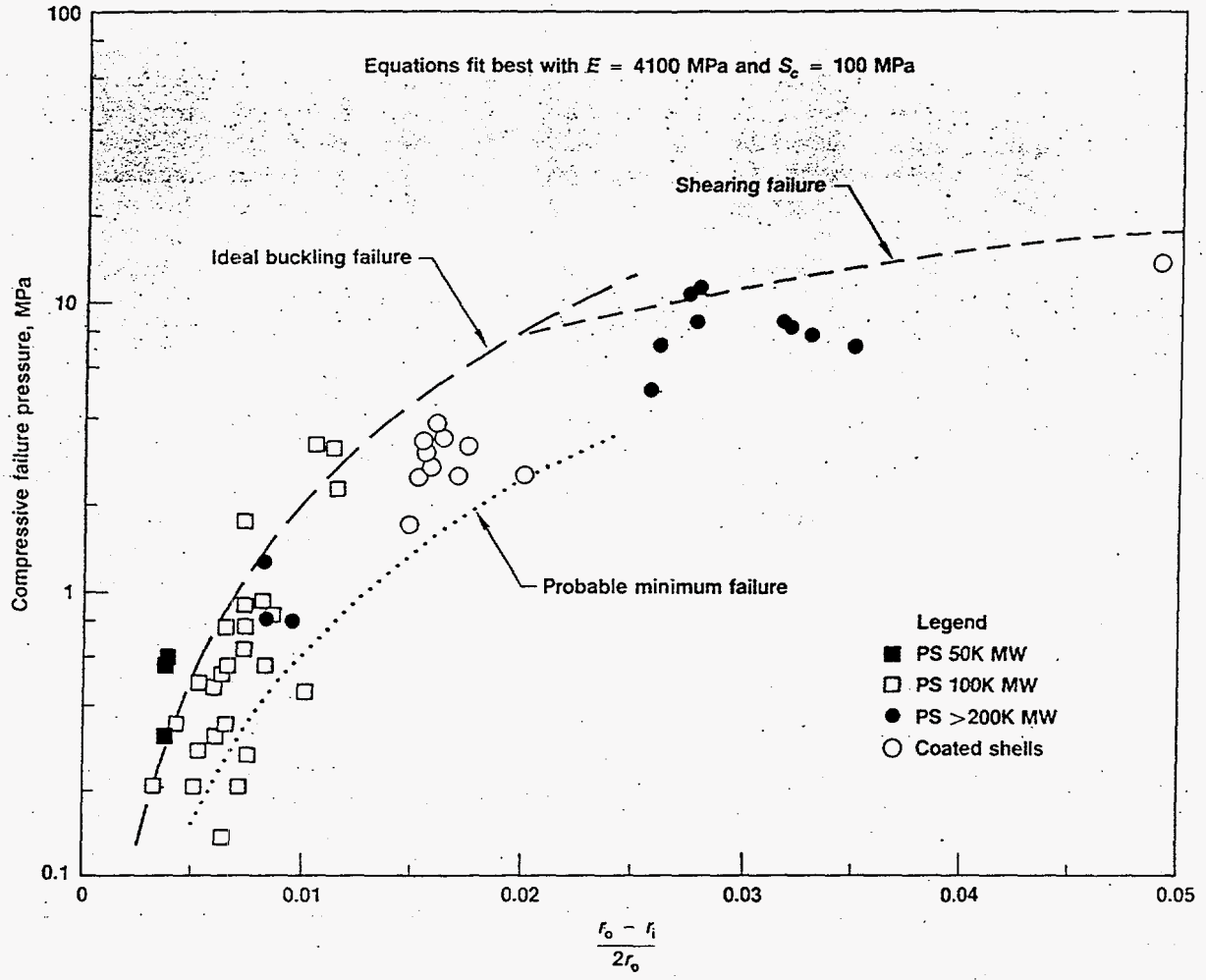
Fig. 6
Apparatus for testing the compressive strength of fuel-capsule shells.



and the wall thicknesses from 1.6 to 30 μm. There is no significant difference in the compressive strengths of the polystyrene shells between those made with different polymer molecular weights or between coated and uncoated shells with the same total wall thickness.

The scatter in the data is probably due to a combination of imperfections in the shells and an inability to accurately measure the wall thickness at the point of failure. For the thin shells, Eqs. (2) and (3) provide an upper and lower bound, respectively, on the failure pressure. As the

(a) Compressive-strength test results.



(b) Buckling failure and shearing failure.

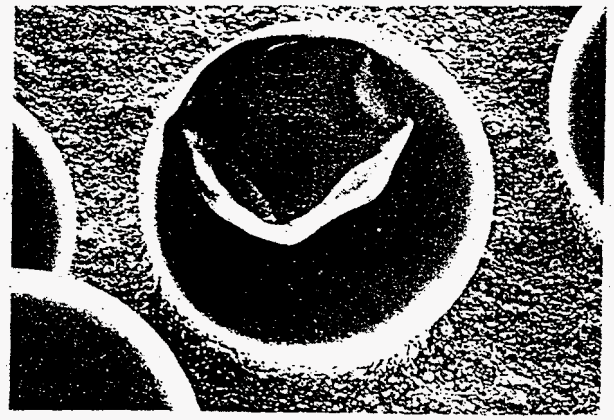


Fig. 7

(a) Results of compressive-strength tests done on the apparatus of Fig. 6. (b) Micrographs showing differences between buckling failure and shearing failure.

thickness increases, the failure mode changes from buckling to shearing, as shown in Fig. 7. The failure pressure of the thicker shells is represented well by Eq. (4). The Young's modulus and compressive strength determined from these experiments are within the range reported for polystyrene.

We were unable to determine the tensile failure by the same video technique because here the failure was often due to a small crack not visible at this resolution. Instead, to test for tensile failure we began by filling the shells slowly to prevent compressive failure, using the results from the permeability and compressive strength tests as a guide. We then quickly released the external pressure and examined the shells by optical and scanning electron microscopy. Some were cooled by liquid nitrogen after filling but before pressure release to determine the cryogenic strength.

The results at room temperature are shown in Fig. 8. The tensile strength of the uncoated polystyrene shells is near 18 MPa, which is about half the literature value for polystyrene. The failure mode depended on shell thickness. The thin shells tended to burst catastrophically, whereas the thick shells tended to develop cracks and spalling near the outside that would gradually grow with repeated testing. Failure pressure of the coated shells was quite variable, although th failure mode was constant in that the polystyrene inner shell would crack but the coating would not. This indicates that the tensile modulus of the coating is less than that of polystyrene, so it stretches too much to provide adequate support. Consequently, the shells coated to Nova thickness are not capable of containing the 20-MPa pressure (200 atm) corresponding to target-capsule fill densities. However, there is some evidence indicating that the coating becomes stiffer with time, perhaps because of cross-linking due to reaction with moisture or oxygen in the air.

Since the capsules are too permeable at room temperature to retain adequate fill gas, they will have to be cooled while awaiting use. Literature data indicates that polymers tend to become both stiffer and

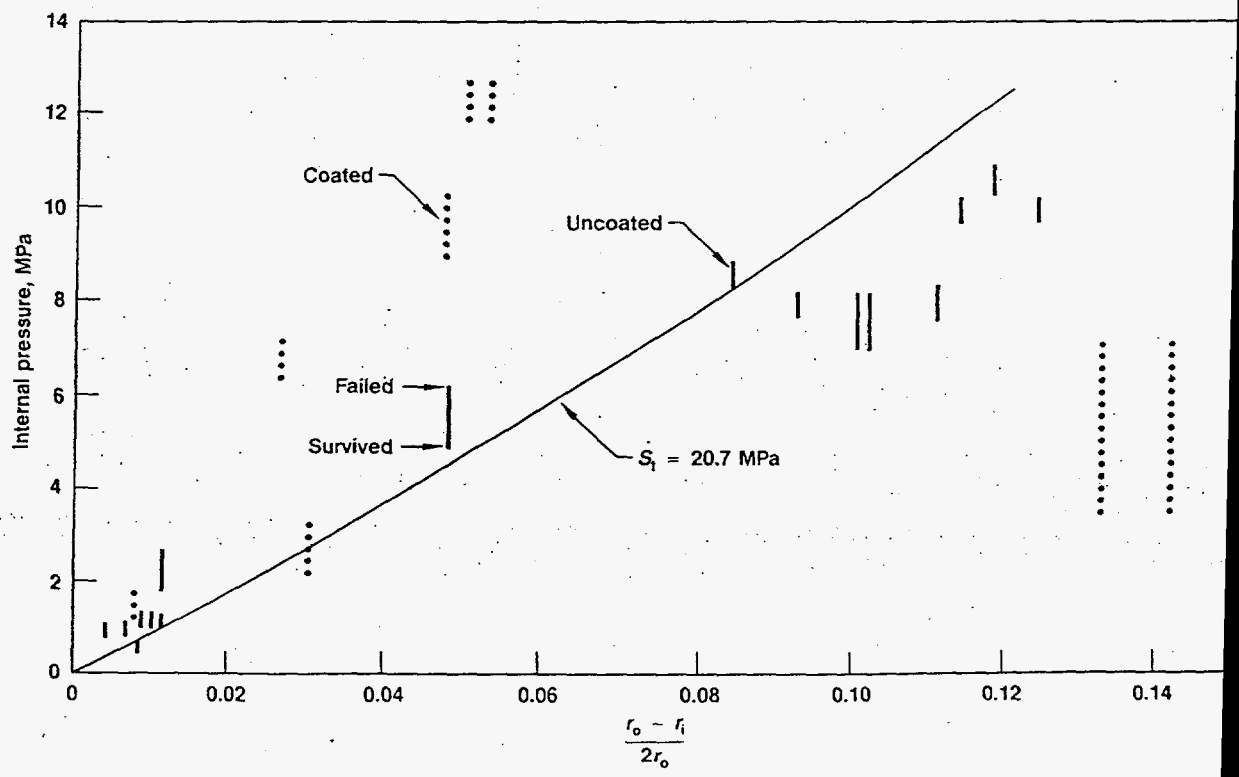


Fig. 8

Results of room-temperature tests of tensile strength of fuel-capsule shells.

stronger when cooled. It was therefore our hope that the CH coating would become stiff enough when cool to adequately support the polystyrene so that the entire wall thickness could be treated as the pressure vessel. At 77 K, we found that the coated and uncoated shells have approximately the same strength for a given total wall thickness, although the polystyrene may be slightly stronger. This indicates that the modulus mismatch that caused the polystyrene inner shell to crack at low pressures at room temperature has largely been eliminated. The tensile strength of the shells at 77 K is about 30 MPa, some 70% stronger than at room temperature. However, we are less certain of the cryogenic value since we could not test very thick capsules because of the limited pressure available.

We are now in a position to better evaluate the filling and maintenance procedures for all-CH-polymer implosion capsules for Nova. We first note that implosion capsules have $(r_o - r_i)/2r_o$ approximately equal to 0.1, so according to Fig. 7 and by use of Eq. (4) they will be able to withstand external pressures as high as 40 MPa. If we allow a safety factor of 5, the maximum pressure drop maintained across the capsule wall during filling should be limited to about 8 MPa. Using permeabilities of 5×10^{-15} and 7×10^{-15} mol·m/m^2·s·Pa for polystyrene and CH coating, respectively, in Eq. (1), we find that the time constant of a polymer capsule 600 μm in diameter by 4 μm thick, coated with 62 μm of plasma-generated CH, is 5 minutes. Therefore, these implosion capsules can easily be filled with D-T to the desired fill density of 40 mg/cm^3 within half an hour using the pressure ramp method and allowing two time constants for equilibration.

The next issue is how far to cool the capsules to maintain fill pressure and prevent tensile failure. Figure 9 shows the temperature dependence of the rate of loss of fill gas for capsules of the same dimensions considered above. We see that the capsule must be cooled to at least 150 K to allow a significant amount of time for handling. At this temperature, the internal pressure has been halved. For a D-T fill density of 40 mg/cm^3, the internal pressure decreases from 20 MPa to 10 MPa, which is greater than the 6-MPa safe working pressure calculated using a tensile strength of 4800 MPa and a safety factor of 4. This result indicates that, to prevent cracking or bursting, the capsule should be cooled to about 90 K, which is significantly lower than the temperature required merely to prevent permeation loss.

As we shift our attention to the capsule design with the PVA interlayer, we need to accurately determine additional permeation and strength properties, including how these properties are affected by the radioactive D-T, to establish fill and maintenance procedures. We can make preliminary estimates based on published physical properties. Again,

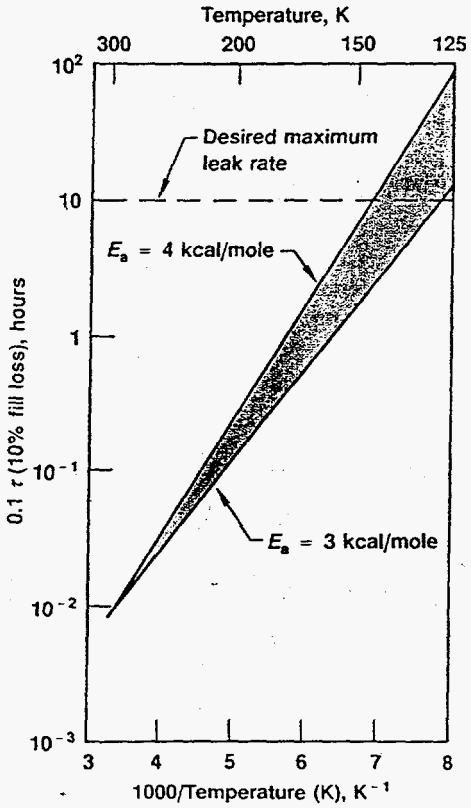


Fig. 9

Temperature dependence of leakage rate of fill gas from polymer fuel capsules.

the fill time depends on the permeability and the compressive strength. In this case, however, the much lower permeability of the PVA causes most of the gas pressure drop to occur across the PVA. Therefore, the CH coating provides support only to the limit of its adhesion. Campbell et al.[7] and workers from KMS Fusion report that CH coating dramatically increases the external pressure that a PVA shell can withstand, so the adhesion must be fairly good. For a PVA thickness of 2 μm and using a PVA permeability of 10^{-18} mol·m/m^2·s·Pa, we find that the permeation half-life of a Nova capsule at room temperature is about 20 hours. Assuming that the compressive strength of the PS-PVA-CH capsule (where PS is polystyrene) is equal to that of a PS-CH capsule of the same total dimensions, we estimate from the calculated half-life that it would take two to three days to fill 40 mg/cm^3 of D-T (20 MPa). This time could probably be reduced significantly if the filling temperature were increased to about 80°C, just below the glass transition of the polystyrene, because the PVA permeability increases 20-fold over that temperature range. PVA is both strong and stiff, so its presence should significantly increase the fill pressure attainable at room temperature. Estimating the tensile strength of a composite shell is difficult, especially when the tensile modulus of the CH coating is so uncertain, but we currently estimate that these capsules should be able to maintain a pressure of more than 10 MPa. Since fill gas leakage is governed by the same time constant mentioned above, we note that significant loss of fill gas will occur over a period of a few hours, so rapid alignment and shooting of the target is necessary. A final pertinent factor, the deterioration of capsule strength and permeability due to D-T exposure, is less well understood and needs to be quantified more thoroughly.

Summary

The desire to obtain fuel capsules with superior implosion and diagnostic characteristics has led us to develop techniques for making and coating polymer shells. We have used the droplet-drying process to produce quantities of target-quality, thin-wall polystyrene shells over the range of diameters required for Nova. We have also demonstrated that the walls of these shells can be made as much thicker as required for the target by a coating process that maintains wall uniformity and smoothness. However, the difficulty in fielding these highly permeable capsules with D-T has caused us to shift our attention to a polymer capsule with an interlayer of PVA, which serves as an adequate permeation barrier. We have developed a simple technique for coating the polystyrene with a thin outer layer of PVA, and initial attempts at further coating these capsules with plasma-polymerized CH coating have been favorable.

Key Words: fuel capsule—glass, polymer, polystyrene; implosion target (for laser fusion); laser fusion; Nova laser facility; plasma polymerization.

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