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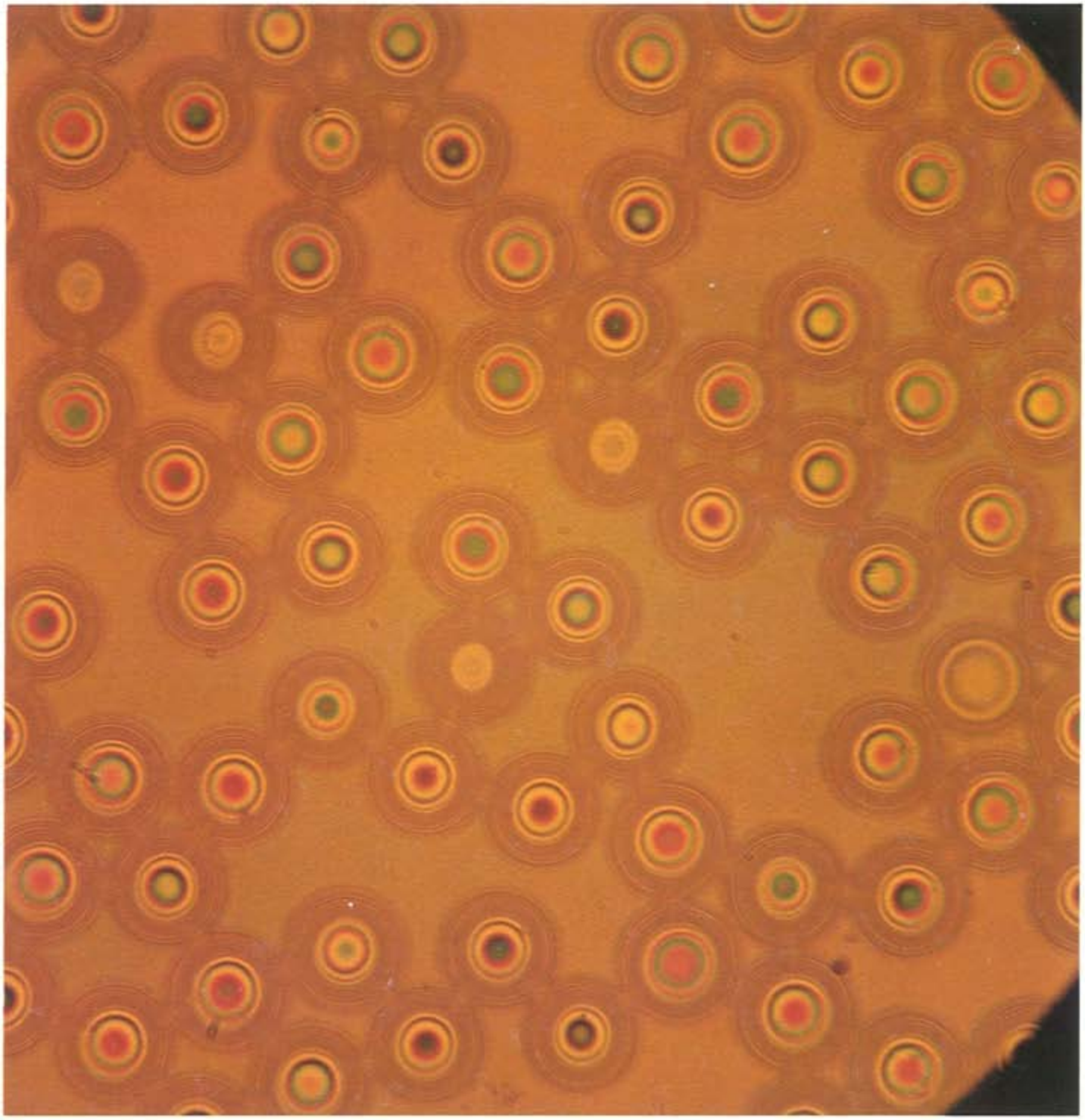

Producing hollow glass microspheres for inertial fusion targets

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A vital component of the inertial confinement fusion program is the ability to produce thin-walled glass microspheres made to exacting specifications with respect to diameter, surface finish, and wall-thickness uniformity. Although we were able to select specimens suitable for our first experiments from large batches of commercially produced microspheres, we needed to develop our own system for producing them for our current and future needs. Instead of rejecting all but 1 in a million, we now have overall acceptance rates of over 90% on many batches of exploding-pusher targets. Furthermore, our precision system enables us to produce microspheres to the much more stringent specifications for the present intermediate to the future high-density ablative targets, for which virtually none of the commercial product is suitable. We have also developed ways to vary the fill gas for diagnostic purposes. We have automated parts of the process, to further reduce costs and increase output.

At LLL and other laboratories throughout the world, experiments are in progress to determine if efficient burning of thermonuclear fuel can be initiated by focused laser beams. When a short pulse of light at extremely high power density strikes a pellet containing deuterium–tritium fuel, the resulting plasma is confined briefly by its own inertia; thermonuclear energy can be released in less time than it takes the pellet to blow apart. For efficient thermonuclear burn, however, the plasma must stay together at extremely high temperatures and densities long enough to let a large fraction of the nuclei react.

For such experiments, a hollow glass microsphere, produced to exacting specifications, contains the fuel. Figure 1 illustrates three such targets: an early "exploding-pusher" target (a), an intermediate-density ablative target (b), and a multishell design still being developed (c). In each of these designs, the innermost layer next to the fuel is a glass microsphere. Glass is an excellent material for this application; it is strong and inert, and meets the specification for a relatively low average atomic number.

A useful property of glass is that fuel gases diffuse readily through even thick glass walls (20 μm or more) at 400°C but, except

through the very thinnest walls (about 1 μm), do not diffuse at all at room temperature. This property enables us to fill most intact microspheres with fuel at high pressure with the assurance that none will leak away before the laser is fired. High-pressure filling also precludes the need for fill tubes or holes that would have to be patched later, destroying the symmetry and uniformity of the wall. Ablative targets used in recent intermediate-density experiments[1] (Fig. 1b) must be filled before the spheres are coated, since the plastic layer is impermeable to fuel gas.

In early inertial confinement fusion (ICF) experiments the target consisted simply of a fuel-filled glass microsphere with an inside diameter of 70 to 100 μm and a wall thickness of about 1 μm. In these "exploding-pusher" experiments the laser beams rapidly heated the glass shell, which exploded and drove a shock wave inward to heat and compress the fuel. The resulting fuel temperatures were high enough to produce easily detectable numbers of fusion neutrons, but fuel densities were only a tiny fraction of that eventually needed for successful application of inertial confinement fusion for commercial power (200 to 2000 g/cm^3, or 1000 to 10 000 times the density of liquid D–T).

Tolerances for the diameter, surface finish, and wall-thickness uniformity of exploding-pusher targets are relatively relaxed, in contrast to those for more recent target designs. For our initial experiments we were able to find suitable glass microspheres by sorting through large batches of commercial microspheres. These are produced by the ton for use in products ranging from bowling-ball cores to

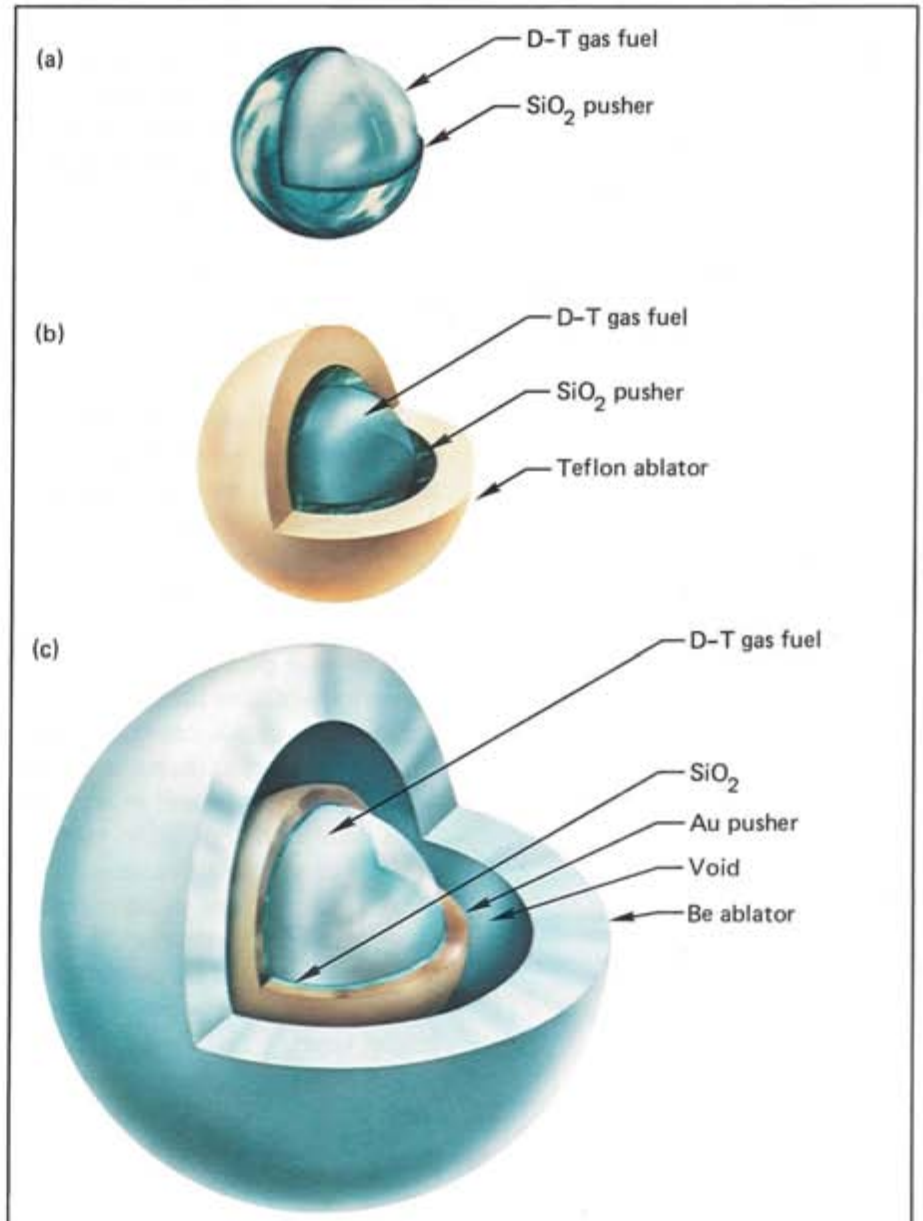

Fig. 1 *Representative ICF target designs. (a) Exploding-pusher target. (b) Intermediate-density ablative target, coated with a layer of fluorocarbon plastic. (c) Multishell design.*

lightweight plastic components for auto bodies.

Such bulk applications impose no particular requirements for uniformity or concentricity. The emphasis is on volume production with little concern for quality. Consequently, only about 1 in a million such microspheres, separable from the rest by laborious screening and inspection procedures, was suitable for exploding-pusher experiments.

The specifications on glass microspheres for ablative ICF targets are very stringent. Acceptable shells must meet a diameter tolerance of $\pm 5\,\mu m$, a thickness tolerance of $\pm 0.5\,\mu m$, a wall-thickness uniformity of $\pm 0.2\,\mu m$, and a surface smoothness tolerance of better than 50 nm. Because the glass microspheres undergo several additional processing steps that have low yields, we also need batches in which at least 80% of the spheres meet all the above tolerances.

Since spheres suitable for ablative targets are not available commercially, we have developed our own processes for producing them in the required sizes and qualities. To do this we needed to understand and control both sphere-formation physics and glass chemistry. Table 1 indicates the improvement we have achieved in microsphere quality over the past three years.

Microsphere production

In the past few years we have developed two separate methods of making hollow glass microspheres: the liquid-droplet method[2] and the dried-gel process. The liquid-droplet process is the more highly developed, reliably producing batches of microspheres 90% of which are within ±10% of the same size and wall thickness. The dried-gel process allows more variation in the glass composition and makes it possible to produce much larger microspheres.

The liquid-droplet process. This process starts with acoustic vibration breaking up a jet of glass-forming compounds in aqueous solution into a stream of uniform droplets. These droplets pass through a long vertical furnace (Fig. 2) where they inflate and fuse into hollow glass microspheres. The average transit time is 1 s. The percentage of solids in the solution, the size of the jet orifice, and the acoustic driving frequency determine the size of the liquid drops and the mass of the resulting microspheres. The furnace temperature profile and air flow, in conjunction with the drop size, determine the diameter, wall

Table 1 *Target-fabrication yields, illustrating the improvement we have been able to achieve by choice of glass composition and control of process parameters.*

	Uncoated			Coated	
	Commercial	LLL		Commercial	LLL
Initial batch[a]	100 000	100 000	**Initial batch**	Suitable	100 000
After sieving for size	10 000	90 000	**After sieving for size**	spheres	90 000
After crush test and D-T load	1 000	60 000	**After crush test and D-T load**	not available	60 000
After optical selection	1	50 000	**After coating with ablative layer**		20 000
			After optical selection		10
Yield	1/100 000	1/2	**Yield**	None	1/10 000

[a]After a 10:1 preselection screening of commercial lots.

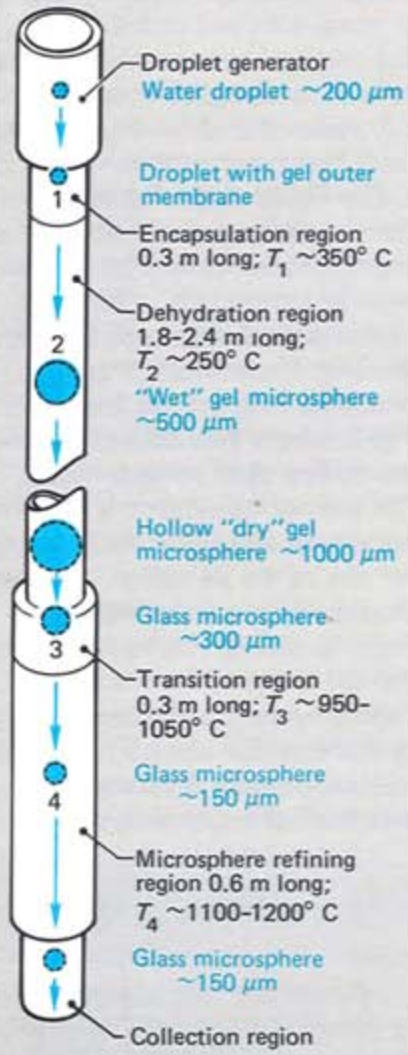

Fig. 2 *The liquid-droplet process for forming hollow glass microspheres. A jet of glass-forming metallic oxides in aqueous solution entering the oven from the top is broken into a stream of droplets of uniform size by acoustic vibration. As the droplets fall through the oven they are first dehydrated, forming porous shells of gelled solids, and then fired to fuse the solids into glass. Control of the process parameters, particularly the temperature profile and air flow in the oven and the composition and size of the droplets, enables us to keep the diameter, wall thickness, and surface finish within acceptable limits.*

thickness, and uniformity of the spheres.

In the upper furnace section the droplets dehydrate and form a gel-like skin on their surface through which the rest of the water diffuses, depositing its solids. The drying temperature is critically important. If it is too low, the droplets dry too slowly, unnecessarily lengthening the required column. If it is too high, excessive vapor pressure ruptures the spheres. When vapor pressure and surface tension are balanced, the result is a hollow, relatively porous microsphere of dried glass-forming compounds.

In the lower, hotter zone of the furnace the gelled compounds melt, react, and fuse into hollow glass spheres. This step in the process depends critically on the choice of glass composition; the glass must have low viscosity to permit surface tension forces to distribute the material into uniform spherical walls. When properly tuned, our system produces batches in which more than 90% of the spheres have the same diameter and wall thickness within ±10% (Fig. 3a). In addition, more than 90% of the spheres within a batch satisfy the exacting surface and symmetry specifications. Figure 3b is an interferogram of a batch of quality spheres that illustrates both the high percentage with uniform walls and the narrow distribution of wall thicknesses.

The liquid-droplet technique has two constraints: the glass-forming compounds must be in solution, and there is an upper limit on the droplet size. Some of the compositions we tried were unsuitable because they formed a gel in the reservoir of the droplet generator, clogging the nozzle. Others became too viscous at normal concentrations or required too large a droplet size when diluted to a workable viscosity; hence, they failed to dry completely in our longest furnace. Thus, to expand the range of compositions and microsphere sizes available to us, we developed a dried-gel process to supplement

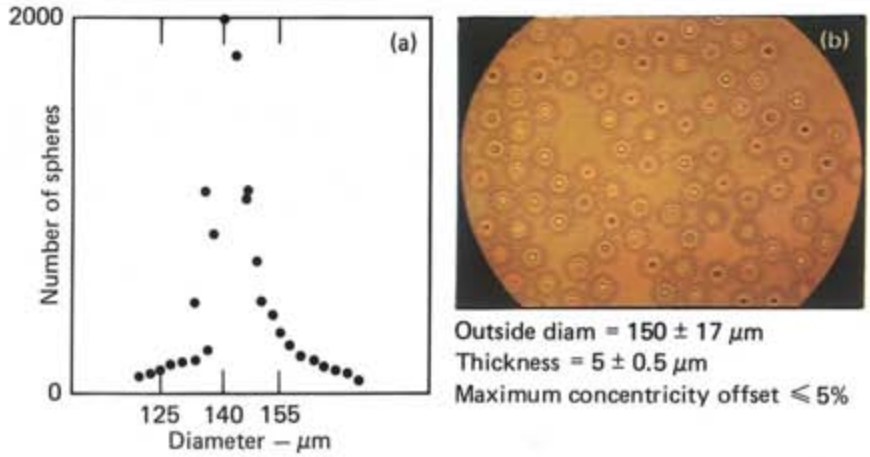

Fig. 3 *Uniformity of microspheres produced at the rate of 100 spheres a second by the liquid-droplet process. (a) Histogram showing the distribution of outside diameters. About 90% of the microspheres have the same diameter within ±10%. (b) Interferogram showing wall-thickness uniformity (80% have the same thickness within ±13%). More than 90% of the spheres satisfy the stringent symmetry requirements for ablative targets.*

the liquid-droplet method of microsphere production.[3,4]

The dried-gel process. In the dried-gel process, we dry a bulk solution of glass-forming oxides and grind the hard residue into fine particles. Sieving selects a batch of roughly uniform particles, which we then pass through a furnace. Trapped water of hydration inflates the particles as the surface starts to melt, forming hollow glass microspheres. The average transit time for this process is about the same as for the liquid-droplet process.

The furnace for the dried-gel process is similar to that for the liquid-droplet method without the upper drying section. This allows us to make the refining section much longer, which enables us to make relatively thick microspheres of high quality. In general, the longer and hotter the furnace, the better the sphericity and concentricity of the microspheres.

Not having to pass the solution through a small orifice gives us greater flexibility in composition, but the necessarily nonuniform particle size produces a wide variation in sphere size (Fig. 4). Furthermore, sieving the product to narrow the size distribution still leaves us with different wall thicknesses (gel particles with different masses can result in spheres with the same diameter). When necessary we density-cut sized microspheres by flotation in various liquids to narrow the wall-thickness distribution. We are also developing ways to produce spherical gel particles that could be sieved before entering the furnace for improved uniformity.

For our initial experiments we used a glass composition similar to that employed in the liquid-droplet system: the main advantage was that we could make more massive spheres (140 μm o.d. with walls 20 μm thick). We can also produce oversize thin-walled spheres (500 μm i.d. with walls 0.5 to 1 μm thick) by adding urea to the gel as an additional blowing agent. Now that we can control the dried-gel process, we are experimenting with lead glass and other high-density glass compositions for the future needs of the ICF program.

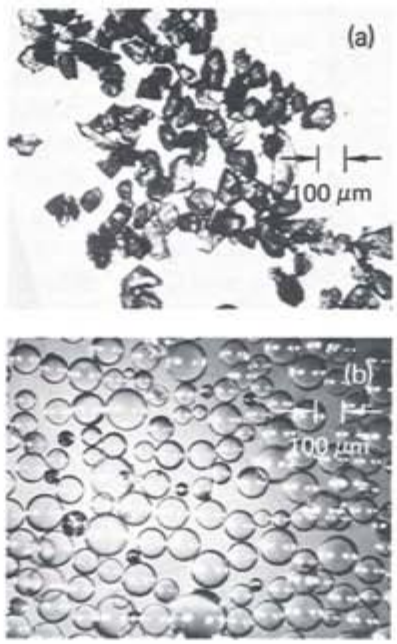

Fig. 4 ***Particles of dried gel (a) and glass microspheres blown from them (b). Although this process produces a wider size distribution than the liquid-droplet method, it permits use of a wider variety of glass formulations.***

Our two microsphere-production processes enable us to make a wide variety of hollow glass shells. Figure 5 illustrates the sizes and wall thicknesses obtainable. For most of our current target designs the microsphere specifications fall outside the commercial supply area on this graph.

Glass composition

Among the various glass-forming compounds, only the alkali silicates are water soluble to any significant degree. Of the alkali silicates, the sodium oxide–silicon dioxide binary system (Na_2O–SiO_2) most easily forms a glass at low temperatures. We chose a composition of about 22% Na_2O and 71% SiO_2 because it is close to the eutectic composition, and thus results in minimum melting temperature. In addition, a eutectic composition reduces the possibility of devitrification (crystallization). This particular glass also has a lower viscosity below 1200°C than other alkali glasses, a vital consideration to ensure high sphericity and concentricity of the microspheres.

However, pure sodium silicate glass is highly susceptible to attack by atmospheric moisture. The primary mechanism appears to be the rapid formation of sodium hydroxide at the surface, which etches the underlying glass. The relatively rapid diffusion of sodium

ions through the glass structure enhances this process.

Accordingly, we alter the Na_2O–SiO_2 system by adding small amounts of network modifiers (potassium and lithium oxides) and the network former beryllium oxide. These modifiers disrupt the continuity of the silicon dioxide network, reducing the sodium-ion mobility and hence the regeneration of the sodium hydroxide surface layer. Their presence greatly increases the durability of the glass and its surface finish.

Surface treatment

Even with network modifiers in the composition, however, the resulting glass still has reactive alkali on the surface, and the sodium ions retain some mobility. It is imperative, therefore, to wash and passivate the microspheres soon after they are formed. Conventional washing with hydrochloric or hydrofluoric acid removes the initial alkali, but the surfaces still deteriorate within a few days on exposure to humid air because of etching by sodium hydroxide that rises to the surface.

We have developed an acid wash procedure that significantly inhibits this etching. The procedure involves several washings with hot 0.5 *N* nitric acid combined with dilute ammonium fluoride, followed by rinses in hot distilled water, acetone, and ethanol. Microprobe analysis reveals that this wash procedure leaches out most of the sodium from the outer layers of the microsphere wall.

Microspheres so treated show no deterioration even after several weeks of exposure to humid air. Figure 6 is a photograph made with the scanning electron microscope of the surface of one of our microspheres. Surface smoothness is much better than the required 200 nm, easily fulfilling the requirements for ICF targets.

Special gas fills

After making glass microspheres of the required perfection, we must fill them with fuel gas, a mixture of deuterium and tritium (D–T). As

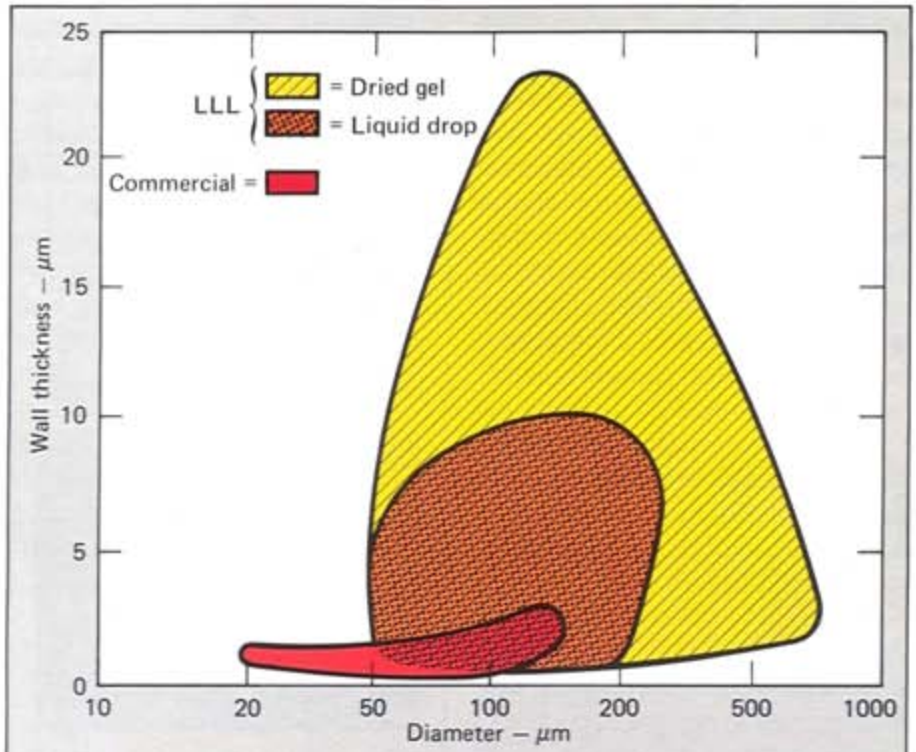

Fig. 5 *Combinations of glass microsphere diameter and wall thickness available from commercial suppliers and from our two alternative processes. For most of our current target designs, the microsphere specifications fall outside the commercial supply area on this graph.*

mentioned earlier, the glass is permeable to such hydrogen isotopes when hot. Filling is a matter of exposing the microspheres to a high pressure of D–T mixture in an oven at 400°C for a day or so.

For diagnostic purposes, however, we would sometimes like to include a partial fill of some other gas. Argon, for example, emits a distinctive x-ray line whose width depends on the density; it can thereby serve as a direct measure of the implosive compression in targets whose glass walls are thin enough to pass the x rays. Bromine, on the other hand, would absorb neutrons from the fusion reaction and become radioactive. If we independently measure the total energy released, we can determine from the amount of bromine radioactivity how dense the fuel had become at the time of nuclear burn.[4]

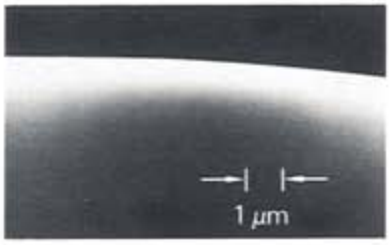

Fig. 6 ***Scanning electron-microscope photograph of the surface of a glass microsphere after being washed with dilute nitric acid to neutralize alkali contamination and to passivate the surface against attack by atmospheric moisture. Wall-uniformity defects are less than 4%; the surface finish is smooth to within 10 to 20 nm.***

Unfortunately, the permeation rate for such gases is a million times less than that for hydrogen isotopes under similar conditions. To introduce them into the microspheres by permeation, we would have to alter the glass composition and then subject the microspheres to high temperatures and pressures for a very long time. Such a procedure would probably be self-defeating, however, devitrifying the glass, roughening the surface by alkali precipitation, and distorting the symmetry because of the prolonged stress.

With our way of making microspheres, however, we can add the nonhydrogen gas during sphere formation.[5] We simply replace the air in the furnace with the gas in question, which diffuses through the walls and remains trapped after the spheres form. On the average, we obtain partial pressures of 20 kPa at room temperature inside our microspheres. We have already used argon gas fills in exploding-pusher experiments and recently developed a way to add bromine to ablative targets.

Ablative targets

The flexibility inherent in these microsphere fabrication techniques has enabled us to develop ablative targets capable of achieving much higher fuel densities. These consist of thicker glass microspheres coated with a layer of fluorocarbon plastic. The laser pulse, which is much longer and may be specially shaped to regulate the appearance of shock waves, burns off the plastic layer at a rapid but controlled rate. The rapid expansion of gases from the ablative layer provides an extended rocketlike inward thrust that compresses both the glass shell and the enclosed, relatively cooler fuel to higher densities.

Because the extended thrust of the ablation produces much higher implosion velocities, it also magnifies the effects of imperfections in the glass microsphere. Hence microspheres for ablative targets must have far more uniform walls and smoother outer surfaces than exploding pushers do. Furthermore, microspheres for ablative targets need much thicker walls than are found in the commercial microspheres. We were able to produce such targets only because we had developed our own microsphere production equipment.

Summary

We have developed our own production facility to provide

hollow glass microspheres meeting the requirements of the ICF program. Control of the production parameters enables us to improve greatly the reproducibility of the process, to vary the composition of the glass and of the fill gas, and to expand the choice of dimensions available. We have also brought under control many of the mechanisms that form surface defects, greatly reducing reject rates. The availability of large batches of target-quality microspheres has made it possible to progress to ablative coated targets, the next phase in our development of ICF targets.

Key words: ablative targets; exploding pushers; glass microspheres; inertial confinement fusion—targets.

Notes and references

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