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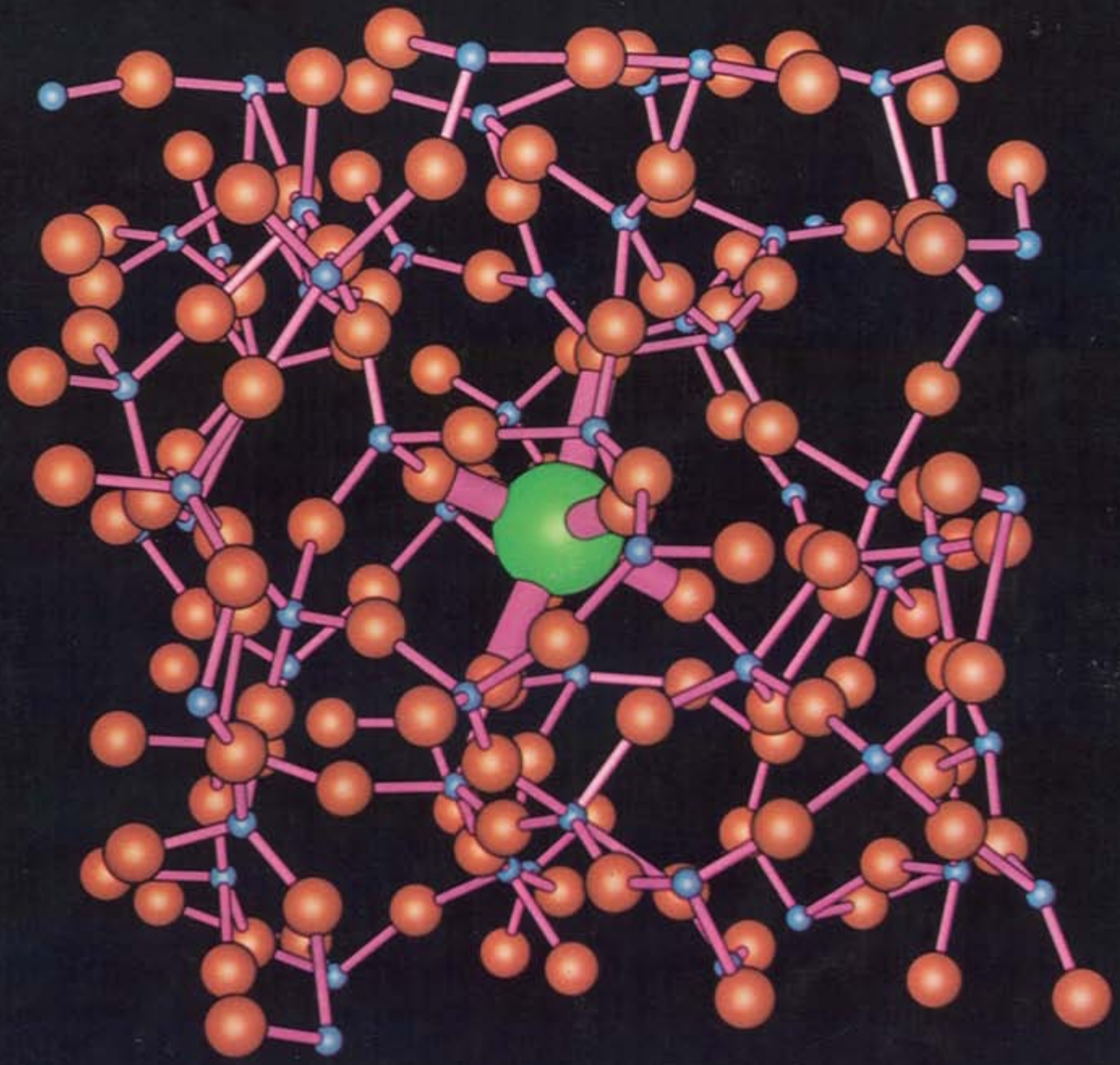

Ultrasmooth coatings for microsphere targets

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Laser-driven inertial confinement fusion research has required us to develop technology for producing high-quality targets that are very well characterized. Current targets consist of glass microspheres filled with D-T fuel and coated with an ultrasmooth layer of ablative polymer. To be acceptable as a target ablator, the coating must have no surface irregularities larger than 0.1 μm, it must be uniform to ±1% of its thickness, and its density must be matched to that of the glass. On glass microspheres from 100 to 340 μm in diameter, we have applied coatings up to 130 μm thick that meet these demanding tolerances. In the two years since the beginning of the project, we have developed several new technologies and produced several hundred target-quality spheres. Our process has set new standards for the quality of plasma-polymerized coatings.

One of the key elements of the laser-driven inertial confinement approach to fusion may be our ability to manufacture ultrasmooth D–T-filled fuel pellets coated with several layers of different materials.[1] Our near-term experiments are designed to help us understand the basic physics of the laser–target interaction. Although large computer codes have been developed that allow us to predict many aspects of target performance, there are still computational difficulties in accurately treating such phenomena as target preheating and Rayleigh–Taylor instabilities, as well as basic questions in the scaling of target interaction physics. Our current experimental program aims to construct a realistic model on which to base our extrapolations to even more efficient target designs.

A supply of targets covering a wide range of parameters is essential for such an experimental program. Since we cannot predict the future demands of the experimental program, our target fabrication methods must be flexible enough to allow us to produce a wide variety of target configurations on demand.

Furthermore, in addition to the constraints placed on the quality of the coating by the target design and by available laser power, the coating process itself must not heat the glass microspheres appreciably. Otherwise the D–T gas will diffuse out through the microsphere walls and be lost.[2] The plasma process we have developed fulfills all of these requirements.

Plasma coating

From a survey of available coating techniques, we determined that plasma polymerization was the best method for producing the required target coatings. Plasma polymers reported in the literature are amorphous and free of voids, important features for laser target

coatings. Plasma polymerization can also produce films having a wide variety of elemental compositions. We have produced plasma-polymerized polyethylene and a Teflon-like fluorocarbon.

In our plasma polymerization process we pass a monomer or precursor gas through a low-pressure discharge region enclosed in a replaceable quartz reaction tube (see Fig. 1). In this region energetic plasma electrons dissociate and ionize the gas to produce a wide range of species. These small fragments of the feed gas recombine to produce long-chain molecules and even macroscopic particles that condense out on all exposed surfaces. Other condensation reactions occur as charged particles and photons from the plasma bombard the coating, breaking bonds and inducing additional reactions. The end result is a glassy light-brown coating.

We generated the plasma for coating the microspheres with a helical resonator (Fig. 1). The rf coil and its coaxial housing comprise a quarter-wave resonant circuit at a frequency of about 10 MHz. This resonant circuit allows the discharge to start reliably over a wide range of plasma power. The

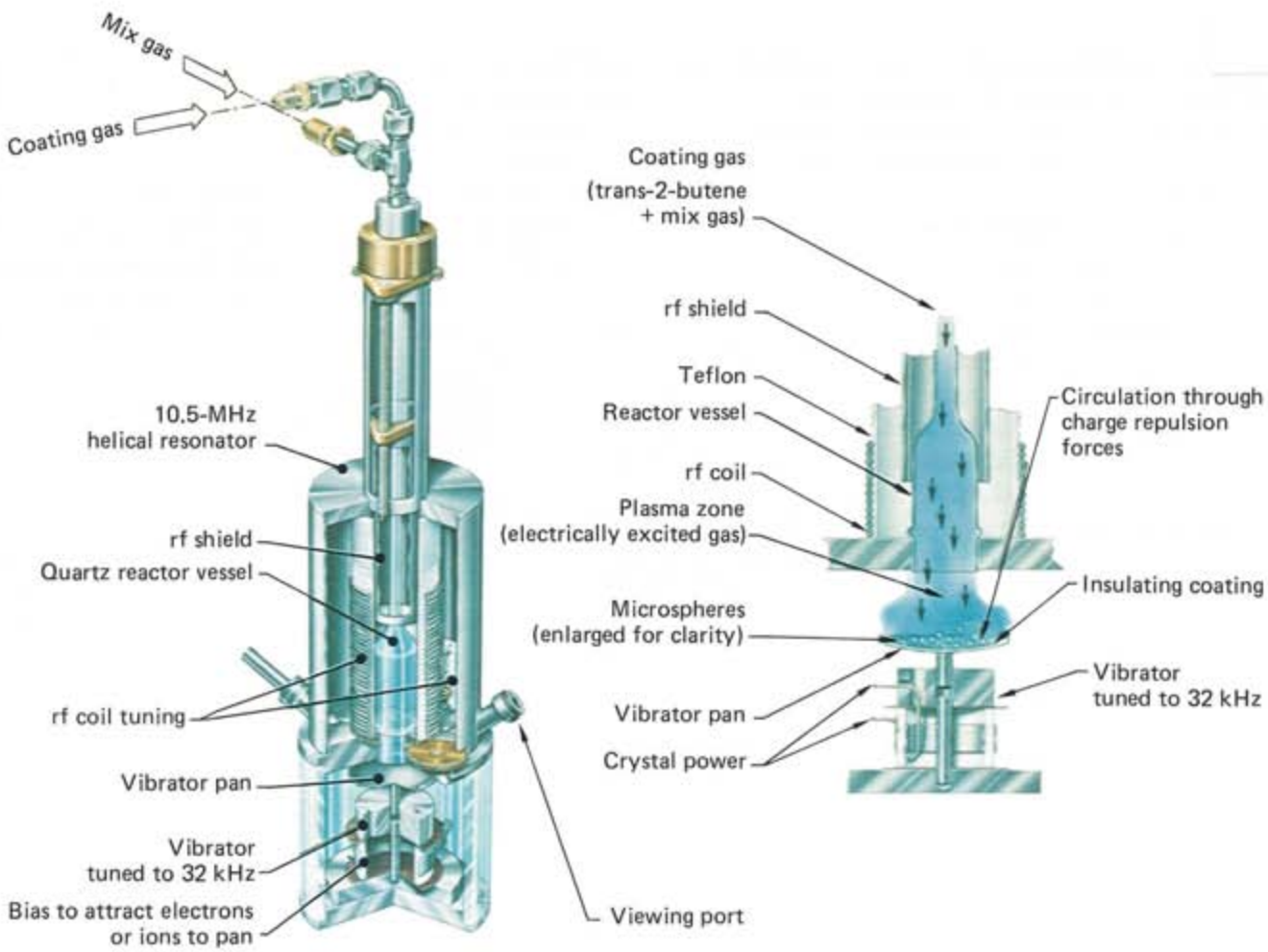

Fig. 1 *Apparatus for coating glass microspheres. The plasma coating process operates by passing an organic gas (trans-2-butene) through a discharge region that breaks the gas into fragments of both ions and radicals. The fragments polymerize on the surface of the microshell to form a uniform ultrasmooth coating.*

plasma is confined to the lower section of the reactor tube and the region immediately above the pan containing the microspheres by a slotted electrostatic shield at the top and by the small diameter of the feed tubes.

To assure the microspheres of random (and hence uniform) exposure to the coating discharge, a piezoelectric transducer vibrates the pan holding them. Biasing the pan to –400 V for 50 μs every 2 ms also stimulates microsphere motion by altering the charge on the microspheres with respect to the discharge. The bias allows spheres whose walls are only 1/70 of their diameter to roll freely in the coating environment.

To begin a coating production run, we thoroughly clean the helical resonator and quartz reaction tube. Once the system is under vacuum, we admit pulses of air through the gas flow path to clear the system of residual particles. We further clean the system with an argon discharge for several minutes and then operate with the monomer gas (but without microspheres) to precoat all the interior surfaces and embed contaminating particles in the initial coating. We then load the spheres into the pan and expose them briefly to an argon discharge to free them of electrostatic charges that would prevent them from rolling freely.

Plasma-coating challenges

The actual coating process begins when we change the feed gas from argon to the organic monomer. From this point on we must monitor the plasma coating environment very closely to assure that it is always optimized for surface finish. A lack of control or a variation in procedure results in surfaces like the one depicted in Fig. 2. Such surfaces are typical of almost any other vacuum coating technique when the coating thickness exceeds a few micrometres. This coating structure is caused by bulk order in the material and the growth of particulate-generated defects.

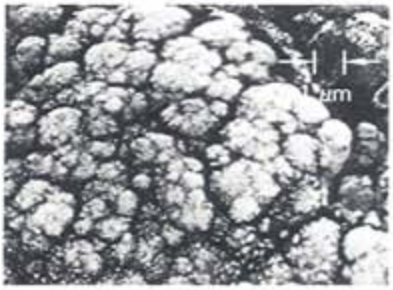

Fig. 2 *The cauliflower-like surface formed under ordinary plasma-polymerization deposition conditions. This surface is far too rough for use on a laser fusion target. We have developed ways to control defect growth so as to produce smooth surfaces.*

In most previous investigations and applications of the plasma polymerization process, surface smoothness was of secondary importance. Consequently, little study had been done on the parameters governing the formation of defects in plasma-polymerized surfaces. To improve the surface finish for our purposes, we needed to be able to identify, understand, and control these parameters. The two principal sources of defects we have identified are process variables and preexisting irregularities on the surface of the microsphere.

Process variables are factors that control the nature of the plasma. They include power dissipated in the discharge, pressure, and rate of monomer flow. Defects arising from such conditions are independent of the nature of the microsphere's surface.

Preexisting irregularities on the surface are also an important source of defects. The relationship between the size of a surface irregularity and the defect it generates is of vital importance in setting cleanliness specifications for the glass microspheres. We have used known irregularities ranging in size from 100 to more than 3000 nm to demonstrate the effect of surface irregularities on coating smoothness.

To illustrate how defects result from surface particulates, we scattered tiny latex beads (1100 nm in diameter) on a clean glass substrate and coated them with various thicknesses of fluorocarbon (Fig. 3). In the foreground, the

coating is less than 1 μm thick, while farther back the coating grows to 20 μm thick. Further inspection shows that the aspect ratio (the ratio of height to width) diminishes as the film thickness increases. This can happen only with an omnidirectional coating flux.

A direct method of minimizing defect growths is to prevent the seed particulates formed in the gas phase from reaching the microsphere surface. These particles are levitated in rf fields, and we can eliminate them by turning the rf power off periodically. We also use ion bias and adjust the power, pressure, and flow to produce a coating free of intrinsic stress; otherwise the coating could fracture and release broken pieces to contaminate other microspheres.

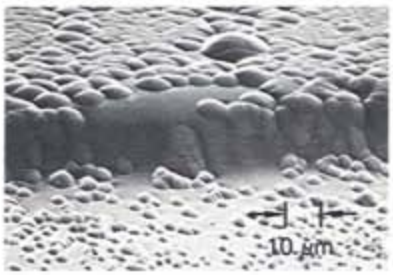

Fig. 3 *Thick and thin layers of fluorocarbon deposited on a glass slide covered with known irregularities, in this case, latex beads 1100 nm in diameter. As the layer thickens, the defects broaden and merge, reducing the aspect ratio (ratio of height above mean surface to width). Similar defects grow from surface irregularities on the glass microspheres; hence extreme cleanliness is essential when thin coatings are being deposited on the microsphere.*

Development of thick hydrocarbon coatings

Future inertial fusion targets may require several layers of different materials coated on the D–T-filled glass microspheres. We have investigated the possibility of making such targets, where the innermost layer is a high density pusher about 5 μm thick. The outer layer must then be a low-density ablator of low atomic number. One option for this ablator is a layer at least 100 μm thick of hydrocarbon polymer with a density of 1.0 g/cm^3. This new target has stimulated our work on production of very thick hydrocarbon coatings.

When we tried to produce hydrocarbon coatings with our old apparatus, we found that coating rates were less than 1 μm/h and that a coating run could span many days. Throughout this time it was necessary for the coating system to operate under precise control to maintain the narrowly defined coating conditions necessary to produce quality targets. To meet this need, therefore, we designed an automated coating system to eliminate human variability from the process.

This new automated coating system, governed by a HP 9825 programmable minicomputer, controls the gas flow into the chamber and the coating chamber pressure. It also provides some basic system functions such as startup or shutdown. It regulates system pressure with a variable conductance valve operated by a computer-controlled stepper motor. It also controls the gas flow rate through the discharge region, monitoring it by measuring the pressure drop across a known conductance.

Since the system is capable of running for many days without an operator, we can make coating runs overnight and through weekends. The coating system also shuts down to protect itself and its product when a problem occurs. The minicomputer that controls the system provides rapid modification of operating conditions with a uniform response, independent of operator, once the best operating conditions are found. It also keeps track of system parameters as a run progresses and prints these out during the run for comparison with other coating runs.

Ultrasmooth thick coatings

The maximum coating rate under which we obtain good surface finish is about 1 μm/h, and it takes several days to reach coatings 100 μm thick. To achieve smooth surfaces, we must control the pressure to ±270 mPa, the flow to ±2.2 μmol/min, and the power to ±2 W.

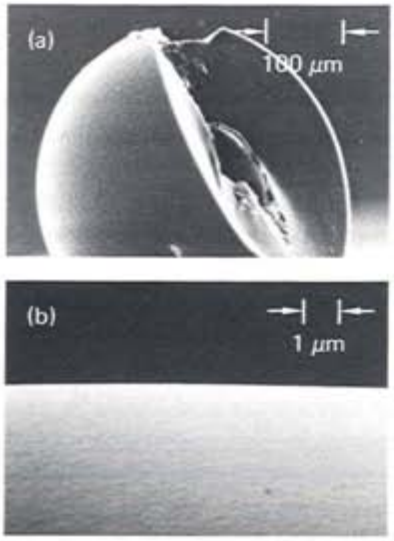

Fig. 4 *Scanning electron microscopy (SEM) provides detailed inspection of microshell coatings. (a) This microshell was fractured to examine the coating structure and to check the thickness. The 110-μm-thick coating shows no internal voids; the bands visible in the coating are caused by interruptions in the coating process. The surface texture (defects 2 μm wide by less than 0.5 μm high) is the roughest acceptable for this coating. (b) Ultrasmooth surface produced by precisely controlled coating conditions. The coating shown has no surface features more than 0.1 μm high.*

We routinely examine many microspheres from each experimental coating run with the scanning electron microscope (SEM) to check the quality of surface finish attained. Rapid analysis of these data enables us to adjust coating conditions before the next run. Figure 4a is an SEM micrograph of a shell coated on a preliminary run in the automatic coater. The chip broken out of its side reveals layers in the coating caused when we interrupted the coating process to check the thickness. The parameters affecting coating rate are much better understood now, enabling us to make spheres of any desired thickness without stopping to measure them. We can now make unlayered coatings up to 130 μm thick with 0.1 μm surface finish (Fig. 4b).

We also radiograph the completed shell in two orientations to determine the concentricity to ±2.0 μm, coating thickness to ±2.0 μm, and glass mandrel thickness to ±1.0 μm. One such microradiograph appears in Fig. 5. We are now working to improve our understanding of this plasma coating process and to find ways to increase the permissible coating rate.

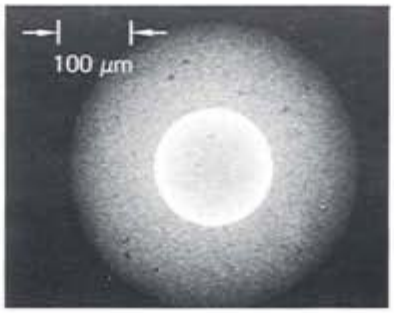

Fig. 5 *Microradiograph of a microsphere coated with a thick hydrocarbon layer. A microdensitometer scan of such a radiograph can provide accurate measurements of coating thickness, glass microsphere thickness, and outside diameter. The microsphere shown has a shell diameter of 155 μm, a shell thickness of 5 μm, and a shell coating of 110 μm of $CH_{1.3}$.*

Future coatings

To demonstrate the adaptability of the plasma process to production of still more sophisticated targets, we have coated multiple layers of materials on glass slides. Figure 6 shows an SEM micrograph of a graded coating on a glass slide, consisting of one layer each of bromocarbon, chlorocarbon and fluorocarbon ($CBr_{0.5}$, $CCl_{1.0}$, $CF_{1.3}$). Such graded layers may be useful in slowing the growth of Rayleigh–Taylor instabilities to

Fig. 6 *A demonstration of multilayer-coating feasibility in which we deposited layers of different densities on a glass substrate. The three layers are bromocarbon ($CBr_{0.5}$) on the bottom, chlorocarbon ($CCl_{1.0}$) in the middle, and fluorocarbon ($CF_{1.3}$) on top.*

improve the symmetry of the implosion and to minimize contamination of the fuel with pusher materials.

The extension of coating technology to very carefully tailored coatings not only is experimentally useful to today's inertial confinement fusion program but also makes the concept of a target factory built around a single coating operation an immediately exploitable goal. We plan further experiments to improve the surface quality of the coatings and the deposition rate of polymer coatings, and to define the practical limits of this plasma technology.

Key words: ablative coatings; electron microscopy, scanning (SEM); microradiography; plasma dissociation; plasma polymerization; surface defects.

Notes and references

1. Previous work on laser fusion targets and techniques for depositing ablative coatings are described in *Energy and Technology Review* for August, 1977 (UCRL-52000-77-8), pp. ii and 5.
2. For recent work on making and characterizing the glass microspheres, see *Energy and Technology Review* for March, 1980 (UCRL-52000-80-3), p. 1.