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FOR LASER-FUSION TARGETS

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ULTRASMooth PLASMA POLYMERIZED COATINGS FOR LASER-FUSION TARGETS*

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Coatings for laser fusion targets were deposited up to 135 μm thick by plasma polymerization onto 140 μm diameter DT filled glass microspheres. Ultrasmooth surfaces (no defect higher than 0.1 μm) were achieved by eliminating particulate contamination. Process generated particles were eliminated by determining the optimum operating conditions of power, gas flow, and pressure, and maintaining these conditions through feedback control. From a study of coating defects grown over known surface irregularities, a quantitative relationship between irregularity size, film thickness, and defect size was determined. This relationship was used to set standards for the maximum microshell surface irregularity tolerable in the production of hydrocarbon or fluorocarbon coated laser fusion targets.

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INTRODUCTION

Plasma polymerization is widely used for coating laser fusion targets because materials can be deposited very uniformly.^{1,2} To meet design standards for ablatively driven targets capable of imploding the DT fuel to high densities, the coating thickness must be uniform within two percent of the wall thickness, void free, and smooth with no defect higher than 0.1 μm . A number of different substrates must also be coated ranging from glass microshells (70 μm to 300 μm in diameter) to microshells coated with copper or gold.³

A problem that exists regardless of the microshell or coating composition is the presence of coating defects. We have found that surface roughness in plasma coating has two basic causes; particle generation in the coating process, and microshell surface irregularities. Particle generation was eliminated by control of the process variables. The second cause of roughness, defects which grow over surface irregularities, do so in a predictable manner. We studied defect growth by analyzing coatings deposited over known irregularities and developed a model which relates coating defect dimensions to surface irregularity dimensions and coating thickness. We can therefore set specific standards for substrate cleanliness to meet laser fusion target specifications.

Plasma Coating Process

Plasma polymerization of various gaseous feedstocks has been studied extensively by other groups.^{4,5} The process of plasma coating utilizes a gas discharge to dissociate gas molecules into ions and free radicals. The molecular fragments later recombine to form a brittle, insoluble polymer coating. Many apparatus geometries, including parallel plate and inductively coupled designs, have been investigated. Our coating apparatus, shown in Figure 1, uses an inductively coupled discharge device operated at 36 MHz to form either fluorocarbon or hydrocarbon coatings. The plasma is formed inside a quartz discharge tube which covers either a piezoelectrically driven vibrator pan that holds several hundred microshells or a molecular beam levitator for coating individual microshells.⁶ The flow reactor configuration allows us to accurately control the residence time of the gas in the discharge zone, which has been found to be critical in eliminating gas phase particle growth.

The parameters that are measured and controlled to give reproducible coatings with the desired surface finishes are the gas flow rates, the total gas pressure in the discharge tube, and the discharge power. Perfluoro-2-butene (PFB) flowed at 0.5 sccm is used as the feed gas for fluorocarbon coating. A mixture of trans-2-butene (T-2-B) and hydrogen at flow rates of 0.3 sccm and 10.0 sccm, respectively, is used for hydrocarbon coating. The optimum system pressure and power for smooth coating is 75 millitorr and 20 watts power. Coating rates are typically

1 $\mu\text{m/hr.}$ making it impractical to manually-control these parameters for the one hundred hours necessary to produce a 100 μm coating. This difficulty was overcome by constructing an automated plasma coater which controls both the total system pressure and the individual gas flow rates. In the event of a system failure, the apparatus automatically disconnects the RF power and DC bias, turns off the gas flows, and closes a valve between the system and the vacuum pump.

RESULTS AND DISCUSSION

Surface Finish Improvement

Early efforts at coating microshells produced surfaces that were completely covered with dome shaped defects. The surface of the target, when examined at 5000X on a scanning electron microscope, resembled a cauliflower. These defective coatings were traced to the presence of small particles that nucleate defects any time they settle on the surface being coated. Two sources of these particles are atmospheric dirt, and particles formed in the polymerization process itself.

Intrusion of atmospheric dirt is minimized by shielding the apparatus when it is open for loading. The quartz discharge tube is cleaned for two hours in an argon discharge to further reduce particulate contamination. A high standard of surface cleanliness on the glass spheres themselves is achieved through a series of washes. The

microshells are washed several times with 0.5N nitric acid and dilute ammonium fluoride. This is followed by rinses with distilled water, acetone, and ethanol. Cleaning microshells in this manner retards deterioration of the surface which is caused by atmospheric storage.

Polymer, of course, forms on any surface in contact with the plasma. An intrinsically stressed coating deposited on the interior walls of the quartz discharge tube is a potential source of particulate contamination. A tensile stress in the deposited coating causes the coating to crack and peel. These flakes contaminate the coater and nucleate defects when they stick to a microsphere. The intrinsic stress is neutralized by controlling the system gas composition. We have found that a combination of trans-2-butene and hydrogen (flowing at 0.3 and 1.9 sccm, respectively) maintained at a total system pressure of 75 millitorr produces a minimum stress coating. For fluorocarbon coating intrinsic stress is not a great problem; a flow rate of 0.5 sccm PFB and a pressure of 75 millitorr is used. An experimental modification which we have found necessary to prevent stress induced flaking for either hydrocarbon or fluorocarbon coatings is to surround the gas discharge section of the plasma coater with a water jacket. This dielectric layer, we believe, reduces the electric field and decreases electron and ion bombardment which may in part be responsible for the intrinsic stress.

Neutralization of intrinsic stress has increased the achievable coating thickness from a maximum of 2 μm (thickness at which the first

flake contamination appears) to over 100 μm , which makes it possible to deposit thick coatings on targets in a single batch run. Figure 2 shows a 140 μm diameter microsphere coated with 110 μm of hydrocarbon that was broken to examine the coating. The coating is uniform and void free, however, the poor surface finish (0.5 μm high defects) rendered this coating useless for laser fusion use.

A second source of particulate contamination arising from the coating process itself is generation of particles in the gas phase.^{7,8,9} These particles, ranging in size from a few hundred angstroms to several microns, are created by gas phase polymerization and are believed to cause the roughness observed in Figure 2. The extent of gas phase polymerization is limited by adjusting the gas composition and residence time in the discharge. Figure 3 shows the effect of T-2-B and hydrogen flow rates on surface smoothness; all coatings were deposited at the same discharge power of 20 watts and pressure of 75 millitorr. Increasing the T-2-B flow rate produces a rougher surface because of increased particle formation. Increased hydrogen flow improves the surface especially at the lowest T-2-B flow because of the reduced residence time in the discharge. The lower rate of particle production at a high hydrogen flow rate and a low T-2-B flow rate results in a smooth surface.

Defect Growth

Even after eliminating all the process sources of particles, we found that defects were still appearing in the microsphere coatings. We suspected the cause of these defects to be irregularities present on the glass microshell substrate. Irregularities arise from degradation of the surface caused by weathering or small particles picked up in handling.

The effect of a surface irregularity on coating quality is shown in Figure 4. The surface quality over most of the microshell's surface is better than $0.1\ \mu\text{m}$. The target quality of the coating, however, was destroyed by a substrate irregularity which when covered by the growing coating became a dome-shaped defect. We now have a clear understanding of the growth behavior of these defects and can predict the aspect ratio of defects based on a knowledge of substrate irregularities. We quantitatively studied the effect of substrate surface irregularities on coating defects. To achieve target quality coatings, it is necessary to both eliminate particles in the coater environment and irregularities larger than $0.1\ \mu\text{m}$ on the glass microshell.

The relationship between substrate irregularities and coating defects was first conclusively proven by studying coatings deposited over diamond etched scratches.¹⁰ Coatings were found to be smooth except over the scratch where a cylindrical bump was found. Later, known surface irregularities (polymer latex beads) were dispersed on a clean glass

substrate. We found that the size of the defect expanded laterally as the coating thickness increased, however, the height remained constant.

If an omnidirectional coating flux is assumed, the topology of defect growth becomes a simple geometric problem. Assuming a hemispherical surface irregularity, successive layers of coating will replicate the original irregularity's contour, and a spherical sector defect will grow concentric with the original irregularity (Figure 5). The locus of the intersection of the defect with the smooth, unaffected surface can then be determined from geometry. Similar geometrical arguments for defect growth were also made for spherical irregularities and defect growth on spherical substrates.

The relationship between a normalized defect radius (R_D/R_I) and a normalized coating thickness (T/R_I) for hemispherical irregularities on flat substrates is

$$\left(\frac{R_D}{R_I}\right)^2 = 1 + 2 \left(\frac{T}{R_I}\right)$$

where R_D is the defect radius, R_I is the radius of the original irregularity, and T is the coating thickness. A similar analysis was performed for spherical irregularities on a flat substrate resulting in the following relationship.

$$\left(\frac{R_D}{R_I}\right)^2 = 4\left(\frac{T}{R_I}\right)$$

Since our main interest is in coatings applied to spherical substrates an analysis of this geometry produced the following relationship for defect growth on spherical substrates of radius R_S .

$$\left(\frac{R_D}{R_I}\right)^2 = 1 + 2\left(\frac{T}{R_I}\right) + \left(\frac{T}{R_S R_I} - \frac{T}{R_S^2}\right) \left(R_I + T\right) - \left(\frac{R_I}{2R_S}\right)^2$$

The equations show that defects grow with parabolic envelopes. The parabolic growth behavior was verified by immersing defected targets in oil and optically observing the subsurface structure of a defect. The model shows that the defect grows laterally as the thickness increases, whereas the height remains constant. Furthermore, the rate of lateral growth is more rapid on spherical substrates than on flat substrates.

To investigate the growth rate of defects and check the model, a range of sizes of latex beads were plasma coated. Figure 6 shows a normal view of the defects resulting from coating over 1.1 μm , 0.3 μm , and 0.1 μm diameter latex spheres. The smallest sphere irregularities produced the smallest defects with the lowest aspect ratio.

The defect size measurements are summarized graphically in Figure 7. The graph plots normalized defect diameter (normalized by the latex sphere diameter) versus normalized coating thickness. Defects resulting from coating over latex particles and microshell substrate irregularities are compared to the growth behavior predicted by the model for spherical and hemispherical irregularities. Target defects were measured on a SEM; both the diameter and height were determined by viewing the profile of the defect. The models accurately fit the experimental points for both latex and target irregularities. The close fit shows that the coating process is well modeled by an omnidirectional coating flux. Having a model that describes the defect growth allows us to decide whether a defect is caused by a substrate irregularity or by contamination produced during coating. This separation aids in identifying and eliminating the problems responsible for defecting coatings and has hastened production of high quality laser fusion targets.

SUMMARY AND CONCLUSIONS

Plasma polymerized hydrocarbon and fluorocarbon coatings up to 100 μm thick were deposited on glass microspheres. A smooth surface finish with no defect larger than 0.1 μm was obtained by eliminating sources of particle contamination and establishing criteria for microshell cleanliness. Gas phase polymerization was reduced by lowering the

coating gas feed rate and reducing the residence time in the discharge zone by the addition of hydrogen. Use of a water jacketed discharge tube and addition of hydrogen neutralized the film's intrinsic stress and eliminated another source of particles.

The growth of coating defects was studied by coating over known surface irregularities. Coating defect dimensions resulting from known irregularities were compared to the predictions of a defect growth model. The model and experiment were found to be in very close agreement. This model contributed significantly to the development of smooth coatings by aiding in the diagnosis of surface roughness and guiding experimental modification.

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Figure Captions

1. Diagram of the plasma coating chamber. The volume of the quartz reactor vessel is approximately 9 cm^3 . The volume of the chamber is 0.9 liter.
2. SEM photomicrograph of a $110 \text{ }\mu\text{m}$ thick coating deposited on a $140 \text{ }\mu\text{m}$ diameter glass microshell. The target was broken to examine the internal morphology of the hydrocarbon coating. The surface texture (defect $2 \text{ }\mu\text{m}$ wide by less than $0.5 \text{ }\mu\text{m}$ high) is unacceptable for ablatively driven targets.
3. The flow rate of both 1-2-B and hydrogen effect the surface finish of the hydrocarbon coating. Increased trans-2-butene flow generates particles through gas phase polymerization and causes surface roughness. Increasing the hydrogen flow decreases residence time and reduces particle formation.
4. SEM photomicrograph of smooth coating with a defect caused by a substrate irregularity. The coating thickness is $64 \text{ }\mu\text{m}$. The defect is $60 \text{ }\mu\text{m}$ in diameter by $8.1 \text{ }\mu\text{m}$ high. Correcting for curvature of the substrate, the irregularity causing the defect is $4.7 \text{ }\mu\text{m}$ high.

5. Coating defects grow by uniformly replicating surface irregularities. The irregularity radius is R_I . The defect radius is R_D . The coating thickness is l .
6. SEM photomicrographs of defects in a 13.8 μm thick fluorocarbon coating caused by polymer latex sphere irregularities. The spherical irregularity diameters are 1.1 μm , 0.3 μm and 0.1 μm .
7. Normalized defect diameter as a function of normalized coating thickness for defects caused by latex irregularities and target defects. Measured defects are compared to the defect growth models for spherical and hemispherical irregularities.

FIGURE 1

CH_{1.3} COATING 110 μm THICK DEPOSITED ON A GLASS MICROSPHERE

100 μm

10 μm

FIGURE 2

EFFECT OF GAS FLOW RATE ON HYDROCARBON COATING SURFACE FINISH

Trans-2-butene
flow (sccm)

0.8

0.5

0.3

0.0

1.9

10.0

Hydrogen flow (sccm)

2 μ m

FIGURE 3

CHARACTERIZATION OF A DEFECT IN AN
ULTRASMOOTH COATING

Coating material $\text{CH}_{1.3}$
Coating thickness $64.3\text{ }\mu\text{m}$
Surface finish $0.2\text{ }\mu\text{m}$

Defect normal view
diameter $60.3\text{ }\mu\text{m}$

Defect height $8.1\text{ }\mu\text{m}$
Sector height $3.4\text{ }\mu\text{m}$
Irregularity height $4.7\text{ }\mu\text{m}$

FIGURE 4

GROWTH OF COATING DEFECTS IN PLASMA POLYMERIZATION

$$R_D = \sqrt{(R_1 + T)^2 - T^2}$$

$$R_D = \sqrt{R_1^2 + 2R_1T}$$

$$\left(\frac{R_D}{R_1}\right)^2 = \left(\frac{1}{\text{Aspect Ratio}}\right)^2 = 1 + 2\frac{T}{R_1}$$

FIGURE 5

COATING DEFECTS NUCLEATED BY KNOWN SURFACE IRREGULARITIES

Coating thickness = 13.8 μm

FIGURE 6

NORMALIZED DEFECT DIAMETER VERSUS NORMALIZED COATING THICKNESS

FIGURE 7