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of Organic Coatings to Metallic and Nonmetallic
Micropellets by the Glow-Discharge Polymerization
of *p*-Xylene**

by

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**LASER-FUSION TARGET FABRICATION: APPLICATION
OF ORGANIC COATINGS TO METALLIC AND NONMETALLIC
MICROPELLETS BY THE GLOW-DISCHARGE POLYMERIZATION
OF *p*-XYLENE**

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ABSTRACT

Laser-fusion targets require thin, uniform organic-film coatings. A coating technique involving glow-discharge polymerization is described for applying highly adherent, extremely uniform, thin films of a high-temperature polymer to a variety of microsubstrates. Polymeric coatings as thick as 10 μm have been successfully deposited on hollow, spherical, 40- to 250- μm -diam micropellets of glass, metal-coated glass, and nickel/manganese alloy. Experimental yields of coatings of a quality acceptable for laser-fusion targets are typically >90%.

INTRODUCTION

In the research on hydrogen-isotope fusion initiated by pulsed laser beams, the lack of sufficiently energetic lasers has imposed a severe limitation on the size of the DT-filled targets. The spherical targets have diameters in the 40- to 250- μm range and several of those being evaluated at Los Alamos are of multishell construction.¹ In such designs the outer shell is often a low-Z material, such as beryllium, boron, or a hydrocarbon.

A hypothetical laser-fusion target is shown in Fig. 1. The inner shell, usually obtained from a commercial source, is a hollow micropellet of glass or nickel/manganese alloy.² The addition of shells to a target of micropellet size is difficult because of the high surface tolerance and wall uniformity required of the shells. The coating technique must be applicable to small numbers (10-100) of micropellets which already have undergone extensive screening and processing. In addition, the micropellets may already be loaded with DT gas. It is important that the organic-coating technique be highly controllable and reproducible so that micropellet losses will be minimal.

Two principal problems set the coating of micropellets apart from the coating of more common objects. First is the difficulty of dealing with objects that are too small to be seen without magnification. Second, the micropellets must be in continuous controlled motion during the coating process to prevent pellet agglomeration and to insure shell uniformity.

The coating material should be of the lowest possible Z number and should have the highest hydrogen to carbon (H/C) ratio consistent with the processing procedure and shell properties. One of the first attempts at applying an organic film to microsubstrates involved the physical vapor deposition (PVD) of polyethylene onto micropellets. Such a procedure was used to apply 1- μm -thick polyethylene films for the Los Alamos Scientific Laboratory (LASL) ball-and-disk target design.³ However, when this technique was modified to coat free-standing micropellets, severe particle agglomeration occurred before substantial coating thicknesses could be achieved.

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Glow-discharge polymerization was then used in an attempt to obtain thicker organic films. Bendix Corp. had done some preliminary work with this process for laser-fusion target applications.² In the glow-discharge polymerization technique, organic films are formed by using an electrical discharge to excite an organic substance in the vapor phase into luminescence. Upon recondensation of the excited monomer, a solid polymer is deposited on exposed surfaces. A unique feature of this system is that when micropellets are placed in the electrical field between the discharge electrodes, they are levitated and oscillate vigorously throughout the coating process, thus contributing to uniform coating. Several organic monomers are potential film-formers,⁴ but *p*-xylene was the monomer of choice because of past experience² and the fact that it is a good film-former and yields a coating with the required low-*Z* number.

EQUIPMENT

A schematic of the coating apparatus is shown in Fig. 2. The vacuum system must be capable of maintaining a pressure of 1 $\mu\text{m Hg}$. The monomer (*p*-xylene) is contained in a glass flask. The electrodes consist of two horizontal, parallel, 1.6-mm-thick anodized aluminum alloy 6061 disks positioned ~ 25 mm apart. In the experiments reported here, electrode diameters range from 25 to 152 mm. A 0.127-mm-thick Mylar film fence taped around the lower electrode on which the micropellets are placed contains the micropellets on the electrode during coating. The power supply consists of an audio oscillator* which drives a McIntosh power amplifier.**

A typical electrode assembly is shown in Fig. 3. To impart additional motion to the micropellets during coating, the assembly can be mounted on an electromagnetically driven vibratory table.

PROCEDURE

The assembled electrodes and substrates are placed into the vacuum chamber and the system is pumped down to a pressure of $<10 \mu\text{m Hg}$. A dynamic flow of argon is then admitted to the system and equilibrated at a pressure of $425 \mu\text{m Hg}$. A 1000-Hz ac voltage is applied to the electrodes, and a glow discharge is formed at 200 V. The glow discharge is maintained in argon for 5 to 10 min to purge the system and to clean the substrate and electrode surfaces. The power is then turned off, the argon flow stopped, and the system allowed to pump down to $<10 \mu\text{m Hg}$. Para-xylene, driven by its own vapor pressure at room temperature, is then admitted and maintained at an equilibrium pressure of $425 \mu\text{m Hg}$. The minimum voltage (400 V) required to maintain a glow discharge with *p*-xylene is applied to the electrodes. The voltage (400 V) and frequency (1000 Hz) are held constant during the coating cycle. Coating thickness is controlled by the length of time during which current is applied to the electrodes. When the desired coating thickness is achieved, power, monomer flow, and vacuum are shut down and the coating chamber is backfilled with argon. The above conditions result in a slow coating rate ($\sim 170 \text{ \AA}/\text{min}$) and the best films.

FILM PROPERTIES

Analytical evaluation⁵ of glow-discharge polymerized *p*-xylene film shows it to be chemically and physically stable. At 300°C there is no softening or melting; in fact, there

*Model 200 CD, Hewlett Packard, Palo Alto, CA.

**Model M1MC 75, McIntosh Audio, Inc., Binghamtown, NY.

is no perceptible physical change. A high degree of spherulitic development, visible under a polarizing microscope, indicates that the film is highly oriented. The infrared spectrum indicates that the basic *p*-xylene structure, including the methyl group, remains intact.

The film has a composition of 82.14% carbon, 7.40% hydrogen, and 10.46% oxygen. The C/H ratio for the monomer *p*-xylene is 11.8, whereas the C/H ratio for the polymeric film is 11.1. This decrease in the C/H ratio after polymerization is caused by the incorporation of oxidation products. The oxygen may originate from impurities (such as water) in the monomer, surface-absorbed impurities in the system, or a vacuum leak. The oxygenated functional groups may be aromatic acids, ketones, aldehydes, or phenolic compounds. They are an integral part of the polymeric structure and probably contribute to the tenacity with which the film adheres to the substrate.

The polymeric coating is insoluble in solvents of widely differing polarities, including water, *N,N*-dimethyl formamide, methanol, ethyl acetate, methyl ethyl ketone, tetrahydrofuran, toluene, carbon tetrachloride, and petroleum ether. Absence of swelling suggests that the material is highly crosslinked. Microscopically, the polymer is uniform and free of voids, bubbles, and gross structural defects.

RESULTS

Successful coating of a variety of microsubstrates has been demonstrated. The coatings are acceptable to LASL theoreticians and experimentalists in the laser-target field. Some typical results of glow-discharge polymerization coating experiments are given in Table I.

A radiomicrograph of a glass micropellet coated with polymerized *p*-xylene is shown in Fig. 4. The 100- μ m-diam glass substrate has a 2- μ m-thick wall represented by the dense inner ring. The polymeric coating is 5 μ m thick and is represented by the less dense outer ring. The uniformity of the coating shows clearly.

ACKNOWLEDGMENTS

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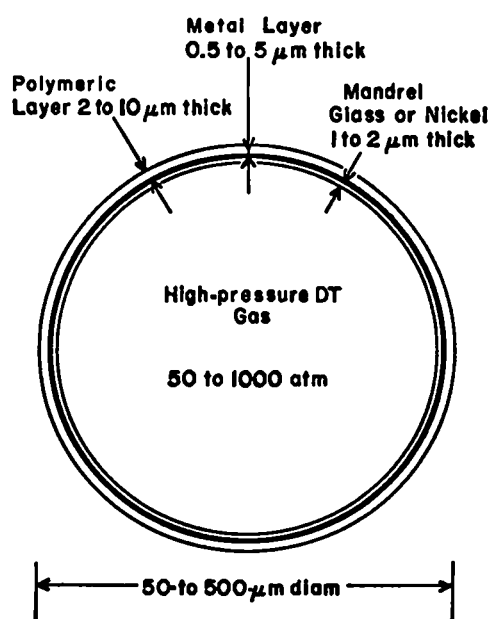


Fig. 1.
Hypothetical laser-fusion target.

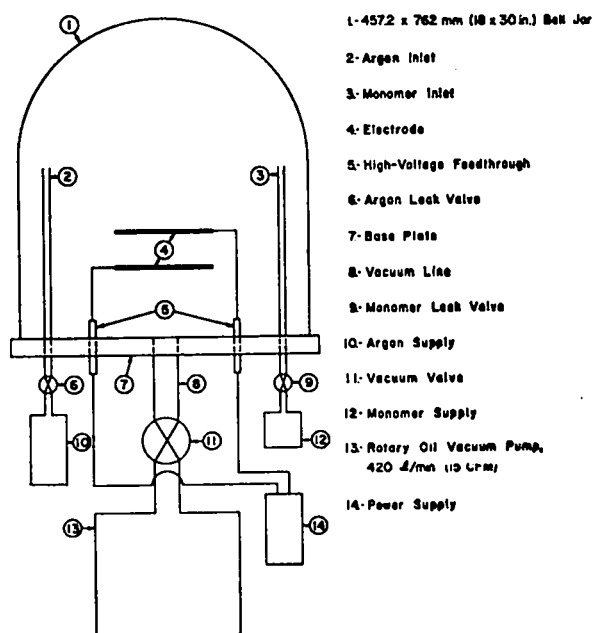


Fig. 2.
Schematic of coating apparatus.

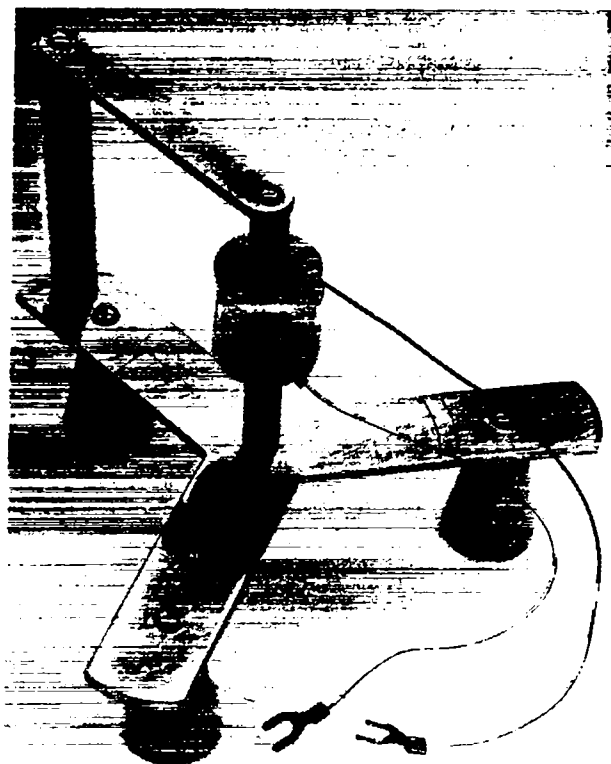


Fig. 3.
Glow-discharge electrode assembly.

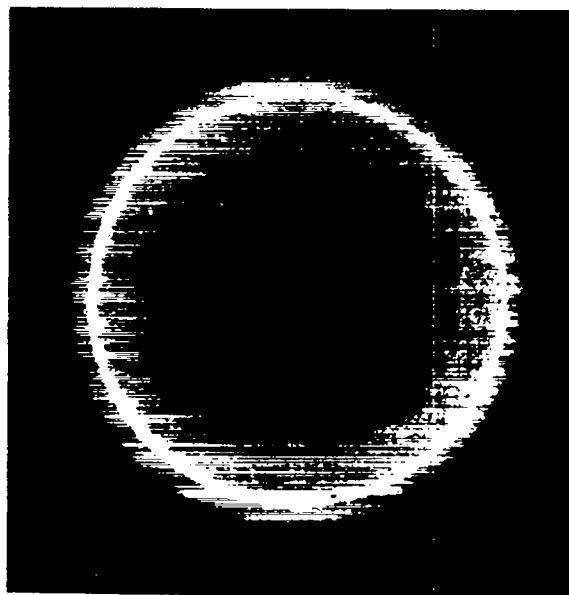


Fig. 4.
Glass micropellet coated with polymerized
p-xylene.

TABLE I
RESULTS OF GLOW-DISCHARGE POLYMERIZATION EXPERIMENTS

Run	Substrate	Monomer Vapor Pressure ($\mu\text{m Hg}$)	Electrode Diameter (mm)	Frequency (Hz)	Voltage (V)	Coating Time (h)	Average Coating Thickness (μm) ^a	Standard Deviation (μm)
GDP-13	3M-P-8593-1 ^b	475	152.4	2000	---	5	10.2	± 0.43
GDP-21	Solacells ^c	425	25.4 ^d	1000	390-400	3	1.3	± 0.37
GDP-24	GNi-19-2 ^e	423	25.4 ^d	1000	400	6	4.4	± 0.68
GDP-25	GNi-19-2 ^e	409-440	50.8 ^d	1000	390	5.5	4.1	± 0.77
GDP-34	3M-Ni-12-20-74 ^f	410	25.4 ^d	1000	400	2	2.8	± 0.32
GDP-35	3M-P-6471-1 ^g	407	25.4 ^d	1000	400	2	3.2	± 0.27

^a Determined from radiomicrographs of arrays of randomly picked samples.

^b Glass microballoons (200- μm diam).

^c Nickel/manganese microballoons (250- to 300- μm diam).

^d Electrodes mounted on an electromagnetic vibrator.

^e CVD nickel-coated glass microballoons (150- μm diam); 2.7- μm -thick nickel coating.

^f CVD nickel-coated glass microballoons (70- to 100- μm diam). Original substrate was 3M-P-6471-6.

^g Glass microballoons (70- to 100- μm diam).