Utilization of polymeric materials in laser fusion target fabrication

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A variety of coating techniques are described that are used for depositing uniform films of a predetermined thickness and high surface quality of a laser fustion target (*L FT*). The first coating method involves the thermal cleavage of di-*p*-xylylene. The product of this thermal treatment polymerizes readily to poly(*p*-xylylene). Addition of pendent groups onto the dimer di-*p*-xylylene extends the usefulness of this coating method. The modified dimers deposit films at significantly higher rates and with improved surface quality compared to films produced from the unmodified dimer. Spherulitic morphology is eliminated and the degree of crystallinity is decreased with the addition of pendent moieties on the phenylene ring. This reduction in the size of the morphological units significantly reduces the surface perturbations caused by these structures, giving rise to target coatings with the required smoothness. Complete elimination of all spherulites and also all lamellae (i.e., crystallinity) occurs with the glow discharge polymerization technique. The glow discharge polymerization of *p*-xylene is used to coat a *L FT* rapidly with a highly crosslinked, amorphous, transparent film having a high surface quality. The system is designed to eliminate powders from depositing on the target's surface. Finally, a technique is described for depositing inorganic and/or organic coatings using dilute aqueous solutions of colloidal particles. This method is unique in that the films are composed of layers of particles with each layer possessing the identical surface charge and chemical makeup.

INTRODUCTION

As fossil fuels become more scarce, worldwide interest will turn towards energy sources such as solar, fission and fusion. Fusion is especially attractive since it offers a vast potential for an environmentally acceptable resource with a virtually unlimited fuel supply[1]. Containing hot plasmas in magnetic 'bottles' (e.g. tokamaks) is one approach to controlling and utilizing fusion power for large scale electricity generation. Another approach uses intense laser pulses to compress, heat and ignite tiny pellets containing the fusile fuel mixture deuterium/tritium (DT). Fusion conditions are achieved when the density of fuel approaches 1000 times conventional solids and the temperature reaches 100 million K. Laser fusion experiments designed to test the feasibility of this approach are being carried out at the University of Rochester's Laboratory for Laser Energetics. This paper describes how polymeric materials and coating techniques are used in the fabrication of laser fusion targets (*LTF*'s) for this program.

A simple and frequently used target is a glass microballoon (typically 100 μm in diameter with a 1 μm wall thickness) containing several atmospheres of an equimolar gaseous mixture of DT. These targets are useful for studying fundamental laser fusion processes, but do not have the potential for large scale energy releases in practical power-producing applications. Higher yield targets designed to reach greater densities with subsequently larger fusion energy outputs will incorporate more complex, structured configurations. An example of such a prototype target design consists of a glass microballoon containing the DT fuel surrounded by a layer of metal and/or polymer. The purpose of the low atomic number polymer coating lies in its ability to ablate as the laser pulse irradiates its surface. This imparts a reaction force to the materials which the polymer layer surrounds, causing the fuel within to be compressed to high density. More advanced targets are multilayer, where the layers are arranged in a double shell configuration separated by a 'void' region. This region can be either a gaseous medium or a very low density foam. The thickness of both the metal and polymer coating is a variable in all configurations. The laser fusion literature describes a broad variety of advanced, structured target configurations[2,3]. One important aspect[4] of these layers is that their surface finish must be smoother than 0.1 μm and conform perfectly to the spherical symmetry of the glass sphere.

Existing polymer coating procedures[5-10], such as solution dipping[5], free radical polymerization[6], and electroinitiated polymerizations[7,8] have been developed, but these methods are not applicable to laser fusion target fabrication. The main problem with these techniques is their inability to coat small spherical particles with the necessary degree of film uniformity and surface smoothness.

In our target fabrication effort, we have developed several techniques for depositing a polymeric coating of a

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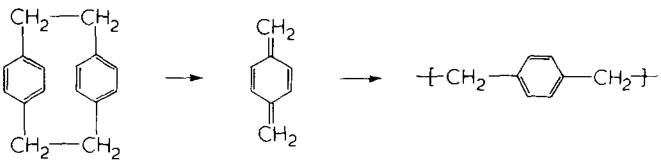
specific thickness with an ultrasmooth surface finish on either a glass or metallic spherical substrate. Three of these techniques will be described. A brief introduction to these methods is given in the following paragraphs.

The vacuum pyrolysis of di-*p*-xylylene (DPX) to form a coherent film of poly-*p*-xylylene (PPX) has been studied extensively[11–15].



DPX PX PPX

The pyrolytic cleavage of the dimer DPX results in the production of the diradical monomer *p*-xylylene (PX). The polymer is formed spontaneously on all surfaces exposed to the gaseous monomer. The film produced is normally pinhole free, but the surface quality can be affected by crystallinity and surface nucleation phenomena resulting in unacceptable coatings for laser fusion targets. Also, the surface quality of the film is dependent upon the partial pressure of the monomer that surrounds the substrate surface. Spherulitic morphology should be avoided at all times because such superstructures will produce surface imperfections that are larger than 1 μm. However, it has been found that spherulitic morphology can be eliminated almost completely by chemically modifying the molecular structure of the starting material (DPX).

The spontaneous polymerization of PX offers certain advantages over other coating techniques. Since the pressure at which this coating procedure takes place is relatively high, (typically 0.0107 kPa), the approach of the monomer to the substrate surface is omnidirectional. As a result, all surfaces, regardless, of their shape or geometry, will be coated uniformly. A second advantage stems from the fact than no intermediate liquid phase exists, since the film is formed directly from the gas phase. Therefore, the effects of gravity and liquid surface tension on the coating uniformity are of no concern.

In glow discharge polymerization (*GDP*), a plasma is produced by electrical excitation in a chamber containing the monomer gas to be polymerized. A film of polymer is formed on all surfaces exposed to the discharge. It is thought[16] that the ions and/or free radicals formed by the bombardment of the plasma electrons with the monomer gas are adsorbed onto a surface, forming a highly crosslinked, amorphous coating.

The importance of *GDP* (with regard to applying a polymeric film to a substrate) lies in the ease of forming pinhole-free coatings[17–23]. Here, however, this coating must conform to precise surface quality requirements. Complete elimination of any structures that cause the film surface finish to deteriorate is important in producing an acceptable polymer coated *LFT*. Since the principal structures produced in *GDP* films are powders[17] formed by molecular recombination in the plasma, it is essential that the formation of such powders be kept from the vicinity of the growing film.

The exacting experimental requirements necessary for the production of films in a parallel plate electrode system have been studied extensively.Shen *et al.*[21] found that low monomer pressures and high ratios of discharge power to gas flow rate, lead to a deposition of powders as well as films. In fact, they observed that there is a transition from discrete powder particles to a coherent film by simply increasing the pressure and monomer flow rate at constant power. A mechanism has been proposed[21] suggesting that the polymerization of the powder takes place totally in the gas phase. These large spherical particles deposit onto the growing film, resulting in a poor surface finish. Therefore, to form smooth, uniform films by *GDP* on *LFT*'s, a system has to be devised by which surface irregularities (i.e. powders) are kept consistently under 0.1 μm and that these particles formed in the gas phase are swept from the system before they land on the substrate surface. It will be shown that a high monomer flow rate is conducive for film formation and that the flowing monomer gas (*p*-xylene) itself can be used to continually sweep out particles from the system.

A technique has been described recently in which coatings of organic and/or inorganic materials could be deposited from a dilute sol onto a substrate. Initially[24], the technique was devised to produce coatings capable of refracting light. In addition, the method has been used to modify the properties of the interfacial region situated between the matrix and reinforcing agent in a composite material[25–26]. This was accomplished by placing the appropriate coating onto the surface of the reinforcing agent before its subsequent placement into the matrix material. This coating technique is generally applicable, for example in the placement of a uniform, high surface quality coating on a *LFT*.

This method is based on the fact that the surface of glass is usually anionically charged in aqueous solutions and that oppositely charged particles (as in colloidal solutions) can be attracted to this surface. A monolayer of particles is formed because further particle deposition is inhibited by electrostatic repulsion. Multiple layers can be formed by placing a layer of oppositely charged particles between each layer of like-charges. Aqueous solutions are the normal deposition media, however, surface tension and gravity essentially have no effect on the quality of the coating, in contrast to conventional solution coating or dipping methods.

The sols used as coating agents for *LFT*'s comprised both organic and inorganic materials[27]. Glassy polystyrene and rubbery polyacrylate are two of the organic materials used, while alumina and silica represent two of the inorganic materials.

EXPERIMENTAL

Target selection, filling and characterization

The procedure for fabricating coated *LFT*'s begins with the selection and characterization of high quality glass microballons. Parameters such as wall thickness, sphericity of microballon, and amount of fuel must be measured accurately and nondestructively for each target. Several methods[28–32] have been developed for the measurement of these quantities: e.g. microradiography, scanning electron microscopy (*SEM*) and interferometry (fringe-shift method).

Before any coating operation, the glass microballoon targets are filled by diffusing equimolar mixtures of deuterium and tritium through the glass wall at temperatures ranging from 350–425°C. At these temperatures, the targets of interest can be filled to virtual equilibrium in a period of less than 20 h. Gases possessing large molecular

Table 1 Effect of pendent groups on the deposition rate of *p*-xylylene and its derivatives

X	Rate (μm h^{-1})*	ΔT (°C)†
H	1	5
Cl	2.5	65
CH_2CH_3	2.5	65
$CH(CH_3)_2$	3.2	105
$(CH_2)_3CH_3$	4.0	115

* The rate of polymer deposition was measured at room temperature in all cases. These rates are only meaningful in terms of the specific geometry of the coating apparatus. The location of the sample is of prime importance in any consideration of experimental geometry

† ΔT is defined as the difference between the threshold temperature and room temperature (25°C). The threshold temperatures were obtained from ref. 11

volume, such as argon, take a prohibitive amount of time for equilibrium to occur. In this case, each microballoon is filled through a laser-drilled micron size hole which is sealed using a polymer plug (one latex particle) of comparable dimension. The plug is melted over the hole and cooled to room temperature to form a seal.

The polymer coatings described in this work were all transparent, so interferometry was used to measure the wall thickness quickly, nondestructively and accurately (±0.1 μm) before and after any coating procedure. The interferometric method measures the optical path through the wall of the microballoon by displacement of white light interference fringes. This displacement of the interference fringes is directly related to the thickness of the target wall. In all cases, the target's sphericity, wall thickness, and surface features, if any, are examined prior to the application of a coating. A target that is not spherical and/or has any defects on its surface, is rejected. After the coating is applied, the target is again examined. If the sphericity or surface quality deteriorated, then this target is also rejected. A Normarsky interference microscope is used for examining target sphericity, coating uniformity and for the measurement of the wall and coating thickness. Also, isolated defects larger than 1 μm can be resolved. Significant increase in magnification (20 000×) and contrast is obtained with *SEM*. Submicron surface features, such as flaws in the polymer coating are easily discerned using this technique.

Coating methods

The preparation of poly-*p*-xylylene involves the thermal cleavage of di-*p*-xylylene. The product of the thermal cleavage polymerizes spontaneously on all surfaces on which the monomer condenses. Due to the high degree of crystallinity of the films and the monomer's propensity for nucleation upon polymerization, large surface perturbations form resulting in unacceptable coatings for fusion targets. It became necessary to modify the symmetrical molecular structure of PPX with the addition of alkyl pendent groups onto the phenylene ring, to lessen the effect of both nucleation and crystallinity.

The substituted paraxylylenes (SPX) were prepared[33] through the addition of alkyl pendent groups onto the phenylene group of DPX by a Friedel Crafts alkylation as shown in *Figure 4*. The same reaction and polymerization sequence was used to prepare other alkyl substituted dimers (*Table 1*) and polymers. *Figure 4* also shows the formation of ethyl-*p*-xylylene followed by vapourization then pyrolysis into two diradical species. These radicals will condense and polymerize onto the surface of a fusion target. The major product is poly(ethyl-*p*-xylylene) but more precisely a random copolymer is formed containing about 15% PPX. Due to the enhanced deposition rate of the substituted diradical species a progressively lower percentage of PPX is expected in the coating as the molecular weight of the pendent group is increased.

The apparatus used in the coating of fusion targets with poly-*p*-xylylene and its derivatives is shown in *Figure 1*. The dimer is placed into the sublimator, and the rate of sublimation of the dimer controlled by varying the temperature in that zone. The gaseous dimer flows through a long quartz tube which is inserted into an electrically heated furnace. It is in this zone that the thermal cleavage of the dimer takes place. The pressure of the monomer gas is measured by a thermocouple vacuum gauge located between the furnace and the collar of the vacuum chamber. Throughout the entire system, all joints are heated to 200°C to avoid condensation and polymerization of the monomer.

The target holder is placed at a right angle to the 'line of sight' of the furnace (*Figure 1*) to prevent heating of the targets. This is important because surface heating significantly reduces the deposition rate of the polymer. The target holder can accommodate 50 targets, whose coating thickness is proportional to the duration of the coating operation. Targets can be taken out of the system at any time and the coating procedure may be resumed easily with the remaining targets.

To ensure that dust and small particles due to both monomer vapour condensation and the vacuum grease do not contaminate the coatings, two 'filters' were included in the system. The first is a plug of glass wool which allows only the dimer vapour to flow into the furnace region. In addition, a circular aluminium baffle is placed in front of the target holder to capture any particles that could strike the surface of the targets.

After loading a weighed sample of dimer into the sublimation zone, and the targets are in place, the system is sealed and evacuated. Air is purged from the system using dry, filtered argon gas. The joints and furnace are brought up to the proper temperature and heat is applied to the sublimation zone with a heat sleeve. The pressure in the system increases until a total pressure of ~0.0094 kPa is reached. Targets can be taken from the holder at predetermined time intervals if different coating thicknesses are required. The system is evacuated again and purged with argon gas. The coating process is continued in this manner until all targets are coated to the desired thickness.

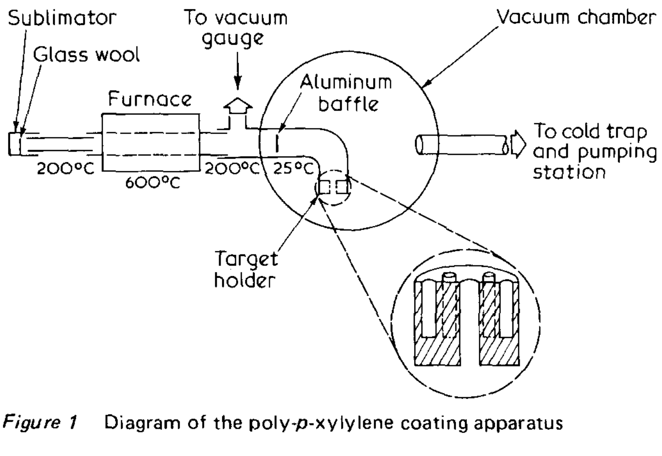


Figure 1 Diagram of the poly-*p*-xylylene coating apparatus

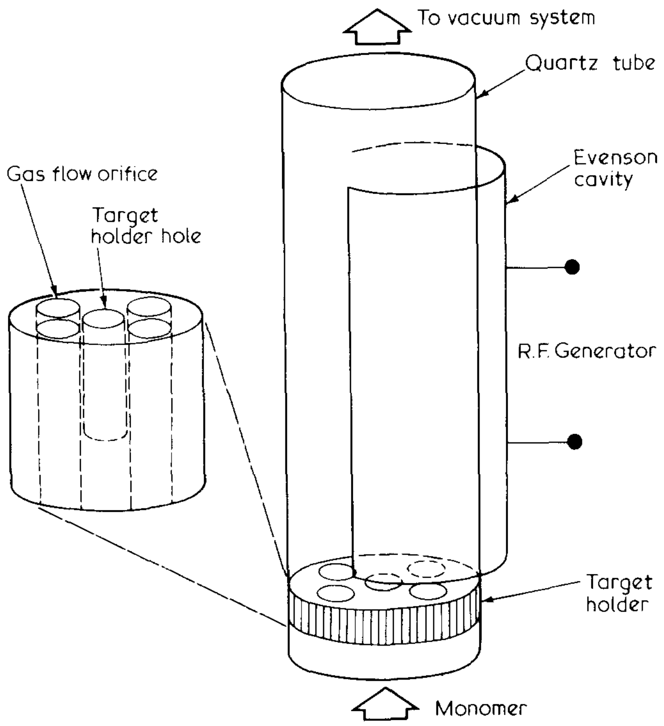


Figure 2 Diagram of the microwave cavity used in the glow discharge polymerization apparatus

An alternative technique for coating *LFT*'s is by glow discharge polymerization. *Figure 2* shows the region in the *GDP* apparatus where the coating of the targets takes place. The target holder, which is inserted into a quartz tube, consists of a hole for holding mounted targets. Each cavity is surrounded by a symmetrical array of holes, allowing fresh monomer (normally *p*-xylene or ethylene) to flow by the target surfaces continually. The stalk (glass fibre) mounted *LFT* is placed in the central hole. The plasma is created in this flowing monomer system by the use of microwave radiation produced by an Evenson cavity[34] – an air cooled, quarterwave coaxial cavity located outside the quartz tube. In this design, the Evenson cavity does not become coated with the plasma polymerized film, therefore the characteristics of this cavity remain constant. The Evenson cavity is powdered by a Raytheon Model PGM-10 microwave generator operating in the 2400–2500 megahertz band at 12.2 cm wavelength.

After placing the fusion target in the target holder and sealing the quartz tube, fresh *p*-xylene monomer is introduced, mixed with a small quantity of air. Oxygen is an essential ingredient in the polymerization process; probably terminating the sporadic and rapid growth of the nodular structures. When the total pressure within the quartz tube is $\sim$0.0402 kPa (0.0368 kPa *p*-xylene/0.0034 kPa air), the plasma polymerization is initiated. This pure gas mixture continually flushes out the plasma region. The duration of the experiment determines the polymer film thickness – normally the coating rate is $\sim$7000 Å min^{-1} which can be varied with changes in monomer flow rate and pressure, or discharge power. For example, an increase in the monomer pressure and discharge power will result in a corresponding increase in the deposition rate. Finally air is introduced into the unit, the quartz tube is detached and the coated targets are evaluated for thickness and uniformity with an optical interferometer.

The third method[24–26] of placing polymer films on fusion targets was used for preparing thick interlayer composite materials and interference films. Starting with a clean glass surface an initial particulate layer is deposited by dipping the *LFT* into an alumina sol (positively charged species), the acidity of which is adjusted to pH 3. The target is removed, rinsed with distilled water and then placed into a solution containing silica or a polymer latex (both negatively charged species). The procedure up to this point constitutes a complete coating step and multilayer coatings are built up by repeating the above mentioned steps. The thickness of the coating is proportional to the number of layers deposited. Coatings have been formed using either silica or polymer latex particles (polystyrene or rubbery polyacrylate copolymer), which are alternated with the positively charged colloidal alumina. Particle diameters of the colloidal alumina, silica and acrylate latex used in this study are 50, 120, and 2000 Å respectively. Since the alumina thickness is negligible compared with the polymer latex diameter, the incremental coating thickness of the polymer films is controlled entirely by the polymer sphere size. Furthermore, the composition of the organic coating is determined by the chemical structure of the polymer, since

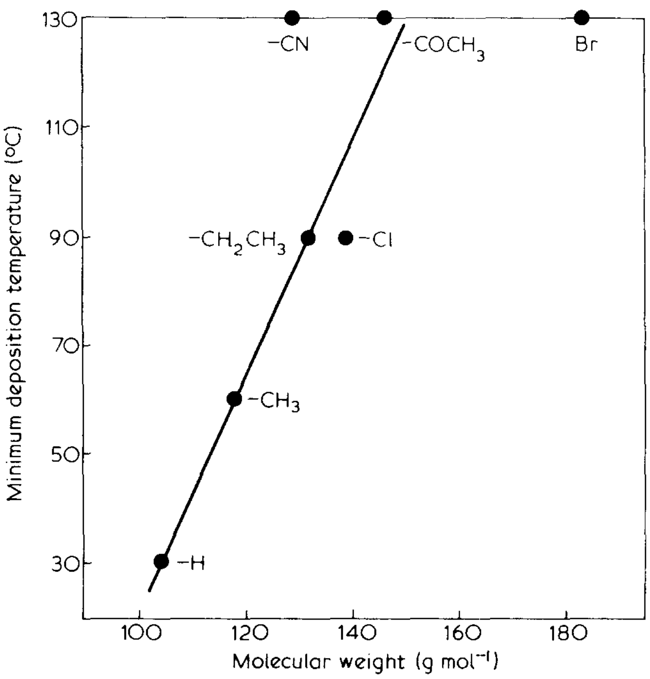


Figure 3 Dependence of the maximum or threshold condensation temperature as a function of the molecular weight of the substituted *p*-xylylene. The condensation temperatures were obtained from ref. 11

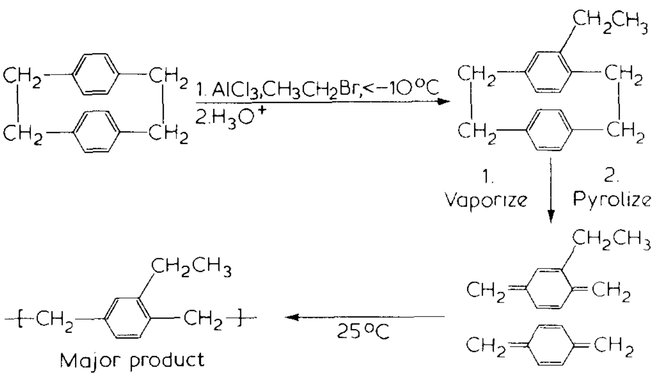


Figure 4 Outline of the Friedel Crafts alkylation of *p*-xylylene and its subsequent thermal cleavage to produce a polymer material

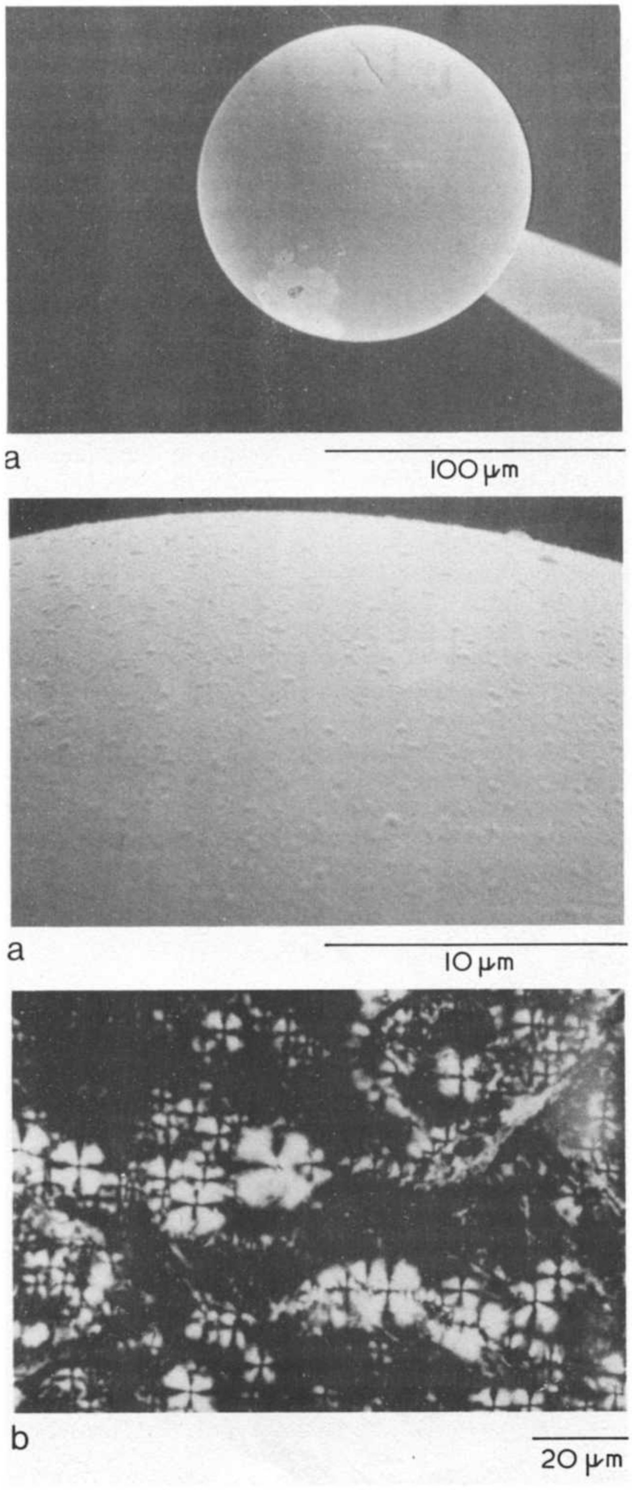


Figure 5 Scanning electron micrographs (a) of the surface of a laser fusion target coated with poly(*p*-xylylene). The morphology of the poly(*p*-xylylene) film is shown in the polarized light micrograph (b)

the alumina serves only as an 'electrostatic glue' to bond the layers of polymer particles. The composition of the inorganic coatings are determined by the chemical structure of both the silica and alumina due to their comparable sizes. The amount of any one component in the coating can be altered by changing its particle size. The inorganic coatings are composed of small particles (50–120 Å) and therefore about 100 layers must be deposited for each micron of coating thickness. To do this an automated programmed dipping apparatus[27] has been constructed to execute a predetermined number of coating cycles. Both supported and unsupported *LFT*'s have been coated by this method.

RESULTS AND DISCUSSION

PPX and substituted PPX coatings

The polymerization and crystallization of the diradical monomer (PX), produced from the thermal cleavage of DPX, occurs simultaneously forming a film composed of folded chain lamellae with occasional spherulitic areas[13]. These morphological entities produce imperfections within the coating and large perturbations on the film surface which significantly reduce the probability of producing an acceptable coating. The problem is how to control the morphology of the films produced by the thermal cleavage of DPX to obtain coatings on *LFT*'s that are deposited rapidly and possess a surface finish that is <0.1 μm. This entails destruction of morphological entities, such as lamellae and spherulites that could perturb the coating surface finish.

The propensity of PPX for crystallization can be reduced through modification of the molecular structure of the DPX dimer. Alkyl groups of varying length (ethyl, isoporpyl and n-butyl) were attached to the phenylene group of the dimer through a Friedel Crafts alkylation. These pendent groups do not significantly affect the mechanism of thermal cleavage, but dramatic changes are observed in both the deposition characteristics of the diradical and in the polymerized material.

Polarized micrographs (at 670 ×) and *SEM* micrographs, were obtained on all coatings, and the results are shown in *Figures 5–8*. The polarizing microscope is used to show the superstructures present in both the PPX and SPX coatings. These micrographs were taken on films polymerized onto a polyvinyl chloride substrate. Because of the small size of a fusion target, it is difficult to obtain a polarized light micrograph of a polymer coating polymerized directly on its surface. The *SEM* is used to examine the surface features of the polymer coatings produced on a *LFT*. For comparison, the pressure at which the polymer deposition took place corresponded to $\sim$0.0104 kPa. SPX coatings were deposited onto the substrates at room temperature and equivalent monomer pressure. The polarized light micrographs show that PPX contains areas with a large concentration of spherulites (*Figure 5*). The dark areas contain crystalline lamellae but these crystallites are not organized into larger structures. Wunderlich observed similar structures in his study of poly(*p*-xylylene) films[13]. The ethyl substituted material is devoid of well-formed spherulitic structures (*Figure 6*) indicating that subtle changes in molecular structure produce significant changes in morphology. Complete elimination of large morphological units is found in both the isopropyl (*Figure 7*) and n-butyl derivatives (*Figure 8*). The *SEM* micrographs show an identical trend in surface quality — large nodules protude from the surface of the PPX coating and to a lesser extent in the poly(ethyl-*p*-xylylene) film, where the surface finish is better than 0.25 μm event though the deposition was performed at more than three times the rate of the former system. An absence of nodules occurs in both the isopropyl and n-butyl polymers. The reduction in large crystalline superstructures correlates directly with the reduction in nodular

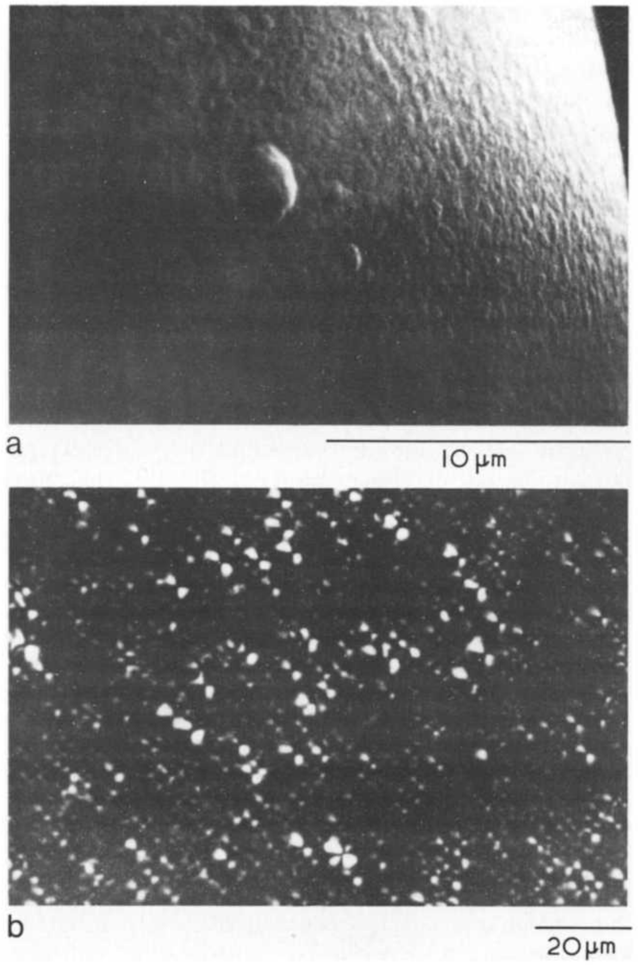


Figure 6 Scanning electron micrograph (a) of the surface of a laser fusion target coated with poly(ethyl-*p*-xylylene). The morphology of the poly(ethyl-*p*-xylylene) film is shown in the polarized light micrograph (b)

growth on the film surface even though the rate of condensation and polymerization was increased.

Further confirmation of the effect of the pendent groups on polymer properties is revealed in differential scanning calorimetry. The melting points (T_m) of PPX and the ethyl, isopropyl substituted PPX are 442, 148 and 64°C respectively. The decrease in T_m correlates directly with the length of alkyl group. Even more revealing is the melting and recrystallization of these three materials. A broad melting range is found for PPX, while no T_m is seen for the ethyl and isopropyl derivative. This indicates that smaller and/or less perfect crystallites are formed upon recrystallization due to the pendent groups inhibiting the crystallization process. No measurable T_m is found for the n-butyl polymer. A direct correlation is noted between the extent of crystallinity and coating surface irregularities on a *LFT*. The accommodation of alkyl groups within the crystalline structure becomes increasingly difficult as the length of the group increases. A point is reached where crystalline structures become exceedingly imperfect and small or cannot form at all. The results shown here confirm that the n-butyl substituted material approximates this situation. In addition, surface imperfections are kept small or completely absent due to the absence of crystallite superstructures. The improvements of the SPX coating relative to the PPX films confirms the necessity of chemically modifying new starting materials.

The increase in length of the pendent group results in enhanced deposition rates, which, in turn, is indirectly related to the substrate temperature. It was found[11] that there is a maximum or threshold condensation temperature (*TCT*) above which polymerization will not take place on a surface at any appreciable rate. This is due to the fact that prior to polymerization the monomer must condense onto the substrate surface. If the substrate temperature is above *TCT*, then there are an insufficient

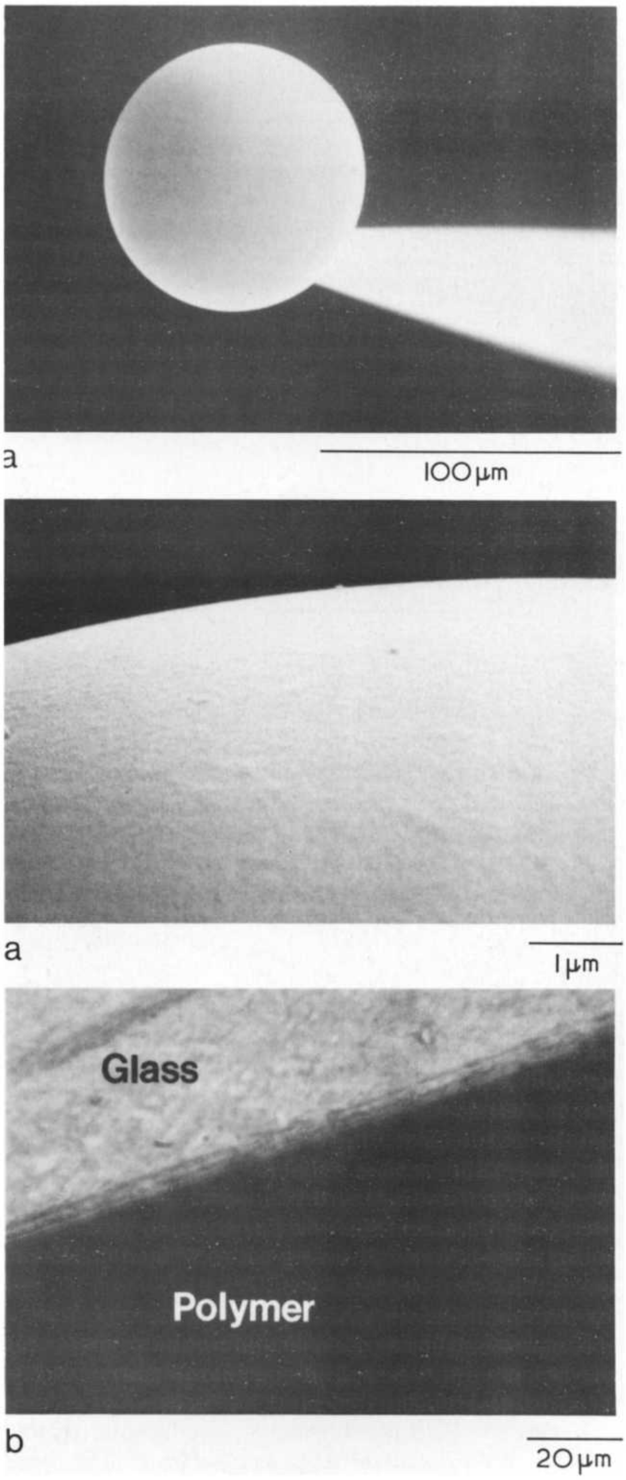


Figure 7 Scanning electron micrograph (a) of the surface of a laser fusion target coated with poly(isopropyl-*p*-xylylene). The morphology of the poly(isopropyl-*p*-xylylene) film is shown in the polarized light micrograph (b)

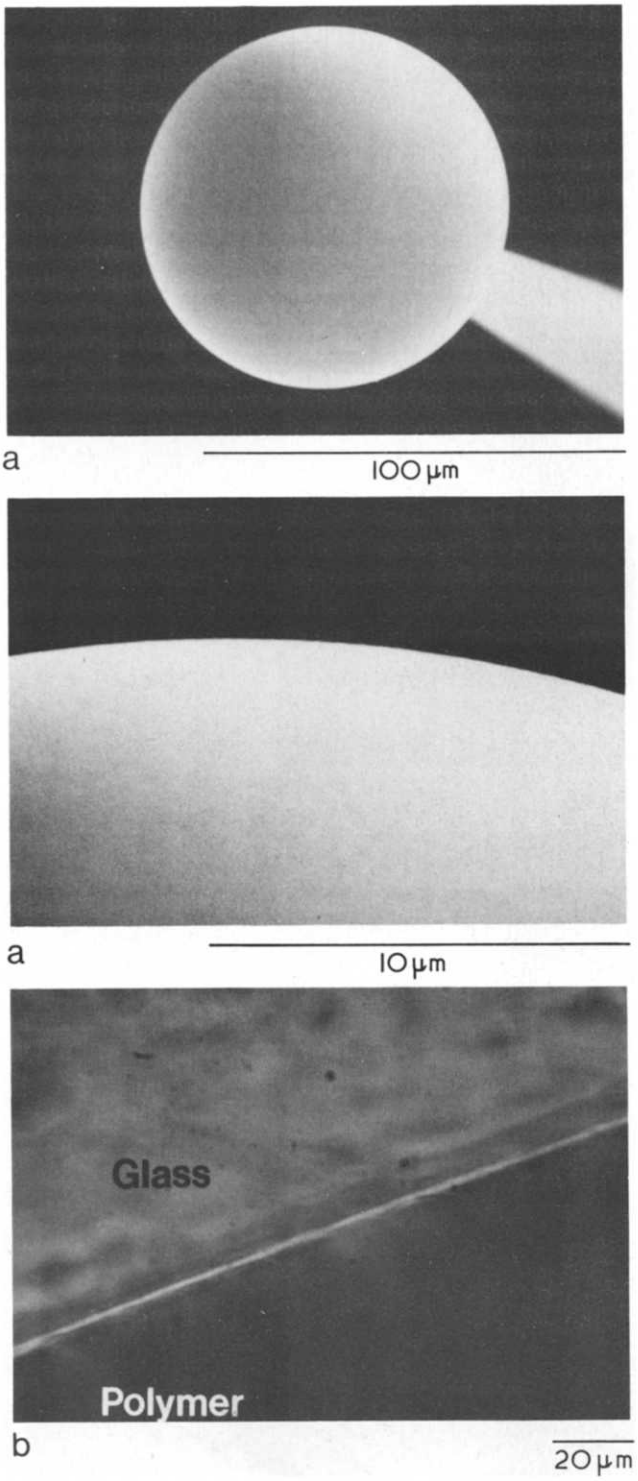


Figure 8 Scanning electron micrograph (a) of the surface of a laser fusion target coated with poly (n-butyl *p*-xylylene). The morphology of the poly (n-butyl *p*-xylylene) film is shown in the polarized light micrograph (b)

number of diradicals condensing, and the deposition rate is negligible. Molecular weight, polarity, molecular geometry, and substrate temperature are the principal factors affecting the rate of condensation. *Figure 3* shows that *TCT* increases with the molecular weight, polarity, and volatility of the diradical. Also, the higher *TCT* is above room temperature, the greater is the SPX deposition rate, as in *Table 1*, where a comparison is made between the deposition rate (measured at room temperature) of four SPX compounds (the deposition rates presented in *Table 1* pertain specifically to the apparatus shown in *Figure 1*.) The absolute deposition rate is a strong function of the distance between the *LFT* and the cleavage zone. When the targets are moved closer to this zone, enhanced deposition rates are observed, for example, if the targets are moved 15 cm closer to the cleavage zone, the isopropyl substituted material will deposit coatings at rates exceeding 25 μm^{-1}. Therefore, the net effect of using an alkyl substituted dimer is that the resultant films are produced at an enhanced rate and are significantly less crystalline in nature. Because of this rate benefit, it is unnecessary to cool the targets deliberately to provide an acceptable coating rate, and the amount of time it takes to coat a *LFT* utilizing SPX becomes considerably shortened. For example, if the distance of the target from the cleavage zone is varied by $\sim$20 cm, the ethyl substituted PX will deposit a film with excellent surface quality at about 25 times the rate as compared to PX. Higher deposition rates occur with the higher molecular weight starting materials.

An alternative means of eliminating crystallinity is to deposit reactive species that immediately crosslink upon polymerization, by using a glow discharge polymerization method.

Glow discharge polymerization of p-*xylene*

To produce uniform polymer coatings on fusion targets by glow discharge polymerization, powder formation must be minimized.

A typical result of *GDP* coating using *p*-xylene as the monomer is presented in *Figure 9*. The conditions in the Evenson cavity (high monomer pressure and flow rate) under which this coating was produced correspond to the transition region between the powder/film and film region. The rate of polymerization was approximately 40 $\mu m/h^{-1}$. Even at this high deposition rate, the coating surface is smooth except for a few hemispherical nodules. At high magnification (20 000 ×), the non-nodular section of the film is featureless.

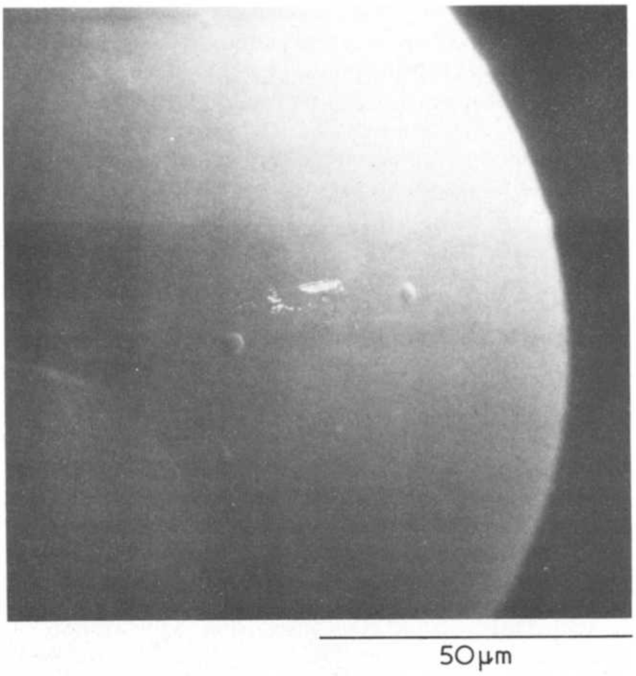


Figure 9 Scanning electron micrograph of a laser fusion target coated by the glow discharge polymerization technique

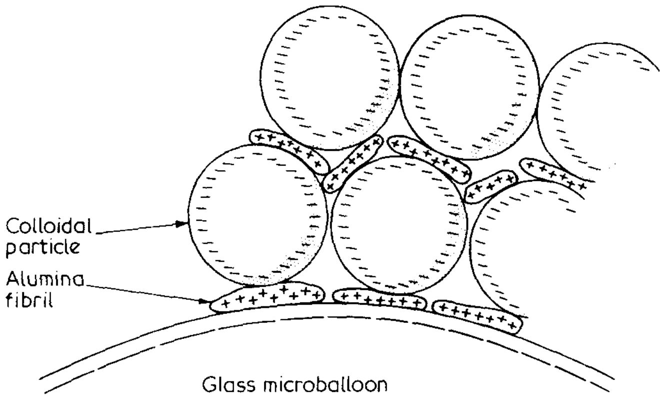


Figure 10 Diagram showing a glass microballoon coated with two layers of colloidal spheres

The formation of powder is assumed to be due to a gas phase polymerization mechanism. After the initiation of the polymerization in the gas phase, the growing polymer material will grow in the gas phase as a powder and/or, in the very early stage of the polymerization, it will strike the substrate surface and become incorporated in a film. The particulates grown in the gas phase are continually swept from the vicinity of the *LFT*'s surface by the monomer's high flow rate. Shen *et al.*[21] have pointed out that the product of the polymerization is dependent upon the rate of polymerization — deposition of film is favoured with low polymerization rates (i.e. high monomer flow rates and high monomer pressure), while at high polymerization rates, powder formation is favoured. These conditions for film production are easy to obtain with the Evenson cavity.

Powder and other particulate material are produced on the surface of the electrodes, such as those used in a parallel plate apparatus. This material can break away from the electrode and become embedded into the growing film causing a rough surface, even though the gas pressure and flow rate are correct. This deposition of material onto the electrode surface is eliminated by placing a barrier between the electrode and gaseous monomer. Also, the growth of particulates in the gas phase and on surfaces can be eliminated to a large extent by allowing fresh gas to flow continuously around the substrate surface (as shown in *Figure 2*). The use of this flow system ensures that the powder is not allowed to build up, and the design of the *GDP* apparatus produces excellent polymer coatings that are both transparent and highly crosslinked.

Colloidal coatings

The polymer film is built up by the addition of individual monomer units, however, an alternative approach[24–26] is to build up a coating with 'macroscopic' particles such as colloidal particles. These particles vary in size from as small as 50 Å to more than a micron. The surface charge of these particles makes them useful as coating agents and can be either negative or positive depending on the functional groups bound to the particle's surface.

As shown in *Figure 10*, the species used to reverse the negative surface charge on glass are positively charged colloidal alumina consisting of fibrils ~50 Å thick. The negatively charged colloidal species deposited on the alumina layer varyed in size and chemical makeup. For most purposes silica, a rubbery polyacrylate and polystyrene particles were used.

Two examples of colloidal coatings on a *LFT* are presented in *Figures 11* and *12*. *Figure 11* shows an inorganic coating composed of alumina and silica particles whose diameters are 50 and 120 Å respectively. A smooth, 1 μm thick film is produced with no perturbations on the surface. Since the T_g of both the silica and alumina is well above room temperature, the film is composed of discrete particles. This is shown clearly by fracturing the coating itself (*Figure 11*), while the particulate nature of the coating species remains intact. To produce a coating that is several microns thick, using particles that are 50 Å in diameter, requires a large number of dipping cycles. An automatic dipping apparatus[27] has been devised in which the immersion times and sequences can be preprogrammed. For example, using this apparatus 5 μm thick films can be produced on a mounted or unmounted (i.e. solution-levitated) *LFT* composed of ~1000 particle layers. A smooth featureless surface is observed when viewed at 10000×.

Figure 12 shows the coating of a *LFT* using a low T_g polymeric material (a polyacrylate). A smooth continuous film is produced because, upon drying, the initially discrete particles coalesce into the film. The surface quality of the film is excellent with only a few small surface distortions (<0.2 μm). These are due to a few particles coalescing in the bulk solution, forming larger particles and subsequently depositing on the substrate surface. In all cases, immersion rates, solution cleanliness, drying procedures, and in some cases, additives in the polymer lattices (surfactants), are critical factors in the production of coatings having a surface finish acceptable for *LFT* fabrication.

A substrate can be coated with any colloidal material (metallic, inorganic or organic) if a surface charge exists, acquisition of which is a relatively common phenomenon, especially with silicates and metal oxides (i.e. alumina and silica). The surfaces of these materials in equilibrium with water are made up of hydrated hydroxide groups. A zeta potential can be imposed which is determined by the H^+ and OH^- concentration (i.e. pH) in the adjacent aqueous solution. The pH at which this potential is zero is defined as the isoelectric point of the surface (*IEPS*), which represents the H^+ and OH^- concentration necessary to suppress ionization of the surface OH groups. At a pH lower than the *IEPS*, the surface is cationic, while at a pH higher than *IEPS*, the surface is anionic. Therefore, by modifying pH, one can alter the charge on that surface. In addition to the use of H^+ and OH^- ions as the potential determining ions, two alternate mechanisms exist for surface charge production. Firstly, the presence of surface functional groups capable of ionization in the presence of a polar solvent is required. These materials are characterized by a surface charge density that is a fixed quantity independent of the concentration of ions in the surrounding liquid. Typical examples include both sulfonated and carboxylated polymers (i.e. polystyrene and polyacrylate) as found in many latex sols. The second mechanism pertains to the crystal lattice of a solid that contains a net negative or positive charge arising from either lattice substitutions or internal defects. The net charge is compensated by an equivalent ionic charge at the surface. In a higher polar solvent, these compensating ions dissociate forming a charged surface.

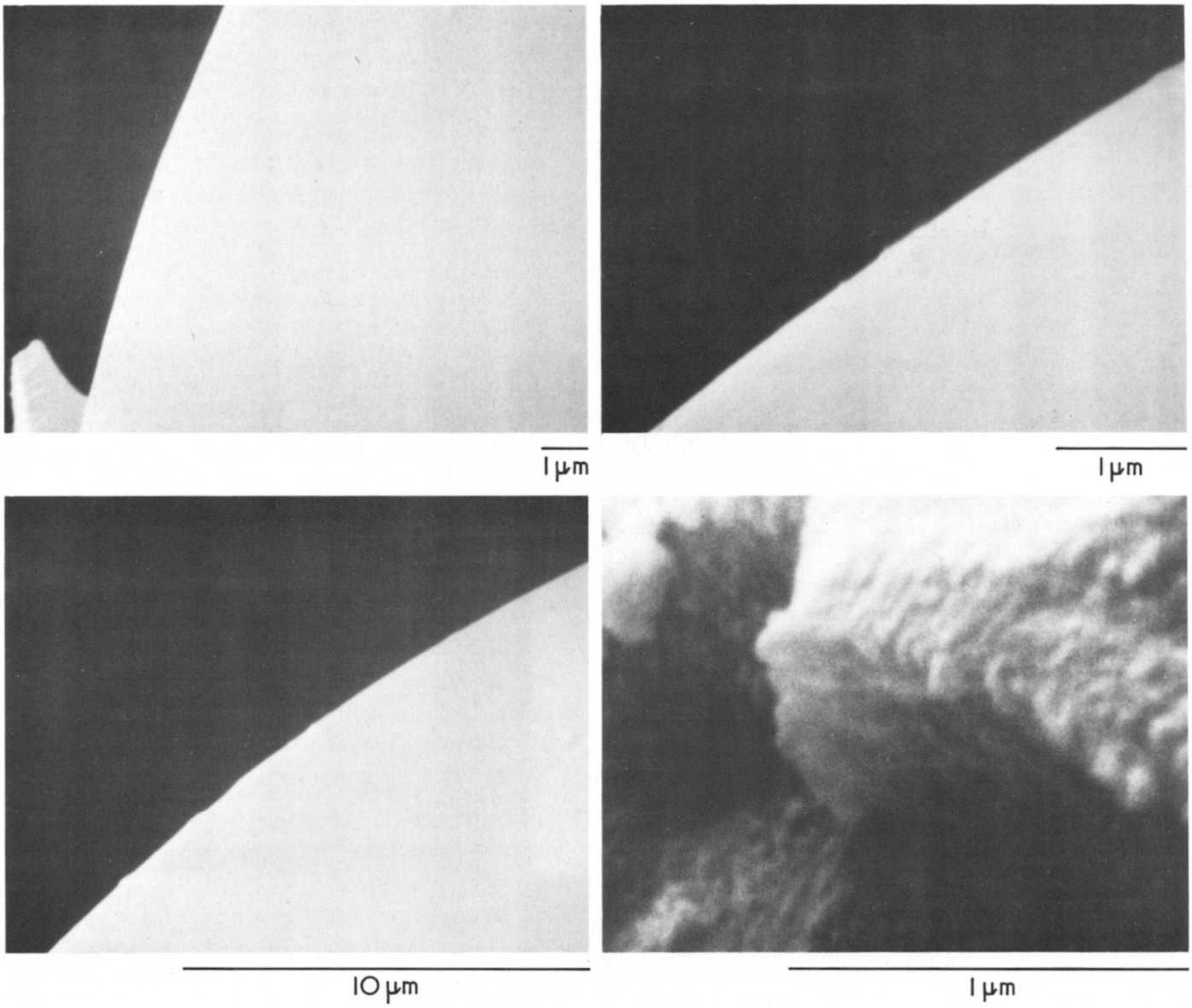


Figure 11 Scanning electron micrographs of a laser fusion target coated with an inorganic film produced by the colloidal coating method

CONCLUSION

A variety of coating methods exist to form a polymer film over a substrate. In most of these techniques, however, the coatings are not of sufficient surface quality to be of value for laser fusion targets, nor can their thicknesses be adquately controlled. The coating requirements for a laser fusion target are so stringent that new techniques needed to be developed for our applications. Additional problems occur from handling and inidividually coating these small, fragile particles.

In this paper several coating methods are presented that meet the requirements for laser fusion target applications. Molecular structure changes of the di-*p*-xylylene dimer increases the rate of deposition and improves the quality of the surface. The chemical nature of the polymer coating also can be varied. Future molecular structure variations will include metallic and/or other nonmetallic pendent groups attached to di-*p*-xylylene, and the number of possible structures is quite large.

Glow discharge polymerization is a rapid method of depositing highly crosslinked, transparent uniform films onto a laser fusion target. The major drawback to its use is the production of powder that originates either in the gas phase or from highly nucleating areas on the substrate surface. Powder formation could be controlled through chemical or physical means, but these methods are not adequate. We have found that by placing the laser fusion target in the flowing monomer and placing the microwave cavity outside the monomer environment, the powder problem is eliminated under a wide range of pressure and flow rates. This arrangement is also compatible with a gas levitation/polymer deposition scheme. In gas levitation, laser fusion targets would be made to 'float' without interparticle interference on a stream of the flowing gas (monomer and air). The target holder, as seen in *Figure 2* would consist of an array of capillary tubes (each tube being about 1 μm in diameter) through which the monomer gas could pass. The flowing gas must impart sufficient force to the laser fustion target for it to be suspended above the capillary array surface. After particle suspension is achieved, the coating procedure would continue as described previously.

The third coating method involves the use of charged colloidal particles (sols) and their inherent attraction to oppositely charged surfaces. Extremely uniform coatings can be deposited on spherical surfaces, such as a laser fusion target, without gas phase or surface nucleation

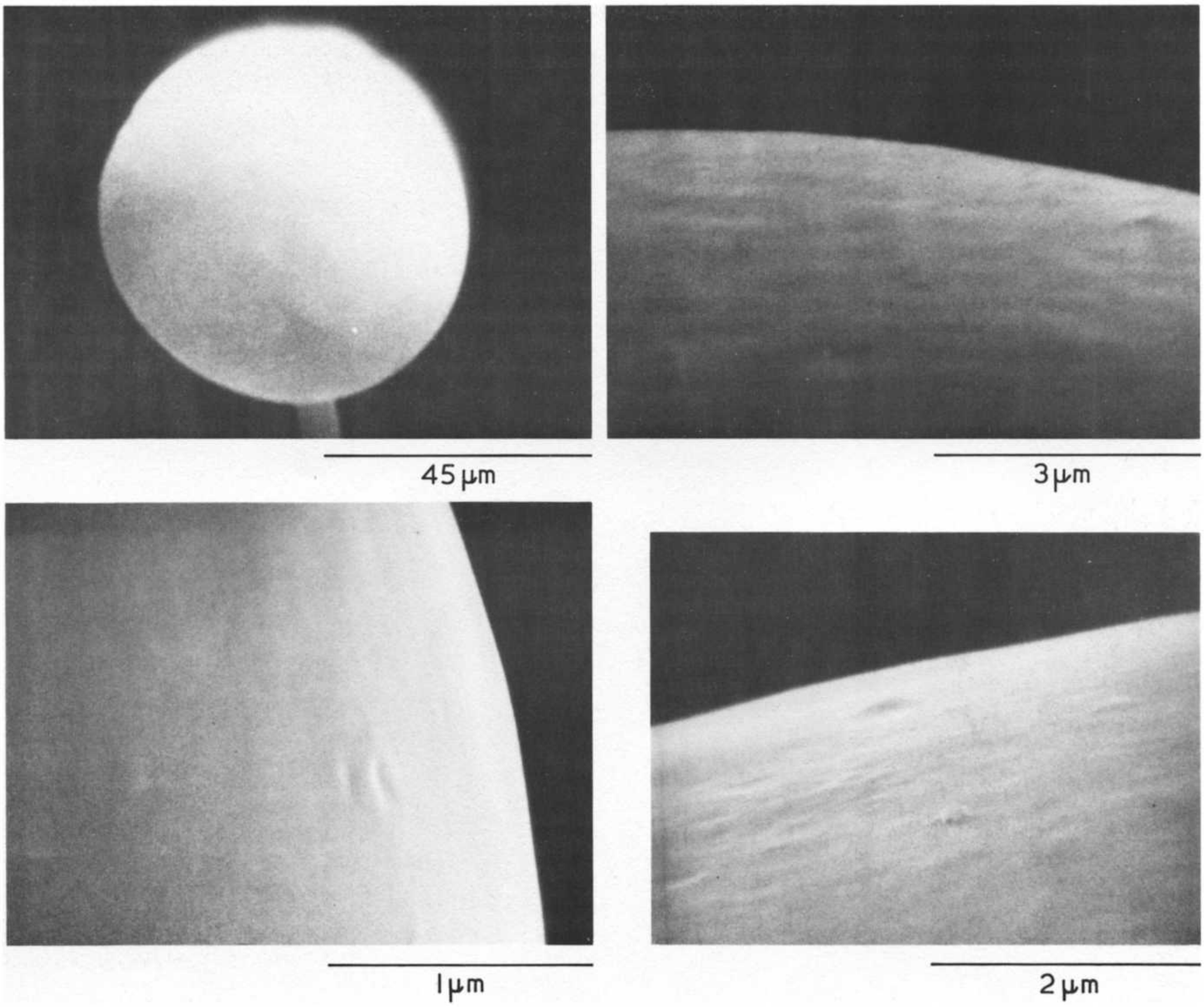


Figure 12 Scanning electron micrographs of a laser fusion target coated with a polymer film produced by the colloidal coating method

problems. The wide applicability of this technique lies in the variety of colloidal systems that exist which can produce films of differing chemical makeup. If the particle can be given a surface charge, preferably through surface functional groups, this method of coating a substrate is applicable. Metals, nonmetals, organic and inorganic materials are representative coating materials. Specific examples are colloidal carbon and clays, water soluble metal oxides, gold sol and organic lattices. In addition, modifications of the substrate surface chemistry through inorganic and/or organic synthetic methods can remove most problems encountered in neutral or only moderately charged surfaces. This technique is readily modified for coating an individual, unmounted laser fusion target.

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