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Initiated chemical vapor deposition polymers for high peak-power laser targets

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ABSTRACT

We report two examples of initiated chemical vapor deposition (iCVD) polymers being developed for use in laser targets for high peak-power laser systems. First, we show that iCVD poly(divinylbenzene) is more photo-oxidatively stable than the plasma polymers currently used in laser targets. Thick layers (10–12 μm) of this highly crosslinked polymer can be deposited with near-zero intrinsic film stress. Second, we show that iCVD epoxy polymers can be crosslinked after deposition to form thin adhesive layers for assembling precision laser targets. The bondlines can be made as thin as $\sim 1 \mu\text{m}$, approximately a factor of 2 thinner than achievable using viscous resin-based adhesives. These bonds can withstand downstream coining and stamping processes.

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1. Introduction

Hot wire initiated chemical vapor deposition (iCVD) of polymeric materials has seen rapid growth as a research field over the last fifteen years. While early studies were focused on elucidating mechanistic details and expanding material sets, recent work has focused on developing applications where iCVD is a critical or an enabling technology. The promise of combining the advantages of chemical vapor deposition (CVD), already an industrially mature material processing technology, with the chemical and mechanical properties of polymers has attracted attention in fields such as sensors, separations, and energy storage/generation [1].

We are developing iCVD polymers for applications in inertial confinement fusion (ICF) and high energy density (HED) science. In ICF and HED experiments, a high peak-power laser pulse generates extreme conditions of shock or pressure at a precision assembly called a *target*, enabling laboratory scale study of material phenomena that occur during nuclear fusion, at high energy densities, or in astrophysical systems. A target must meet exacting mechanical, dimensional, and chemical specifications in order to provide suitable data for ICF and HED

researchers [2]. This paper reports on iCVD materials intended to address needs in the fabrication of these laser targets.

One of the key target components for ICF experiments is the *ablator*, which is a spherical shell that implodes symmetrically to compress and fuse hydrogen isotopes contained within the shell [3]. Plastic ablators are currently fabricated via plasma CVD, whose low intrinsic deposition stress enables them to be coated in the thick layers ($\sim 200 \mu\text{m}$) required for implosion experiments while still taking advantage of the material purity and low surface roughness enabled by CVD. However, defects introduced during the plasma-synthesis of the ablator cause them to photo-oxidize rapidly under visible light [4]; the presence of oxygen in the ablator degrades its implosion performance during ICF experiments [5]. Here, we report on the use of the non-plasma iCVD process to synthesize transparent and photostable poly(divinylbenzene) (PDVB). We have also characterized the intrinsic film stress to understand the potential for growing the thick layers required for ablators.

HED experiments often study the propagation of shock from one material to another and targets consequently include two bonded metal foils. Currently, foils are bonded using commercial adhesives that are applied as viscous resins and cured as a solid polymer. The presence of the adhesive layer between foils can perturb the shock as it propagates from one metal foil to the other. While careful application of glues can yield bonds as thin as $2 \mu\text{m}$, future targets will require bonds that are submicron in thickness. A number of studies have reported the iCVD deposition of adhesive polymers [6]. This approach is attractive because the thickness of the adhesive layer is determined by the

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thickness of the deposited polymer layer. Here, we extend this work to bonding metal foils using thin layers of poly(glycidyl methacrylate) (PGMA) and investigate the survivability of these bonds to downstream processes such as coining and stamping the foils.

2. Experimental details

2.1. Polymer deposition

Deposition of plasma polymer was conducted in a custom-built RF-plasma chamber described in full detail elsewhere [7]. *Trans*-2-butene (95%, Matheson Tri-Gas) and hydrogen flowed into the chamber (0.1 and 10 sccm, respectively) and films were grown at an RF frequency of 43 MHz and plasma-power of 13.5 W. iCVD films were deposited in a commercial chamber (GVD Corporation) equipped with a Nichrome filament (Goodfellow). Divinylbenzene (DVB, 80%, Sigma-Aldrich), glycidyl methacrylate (GMA, 97%, Sigma-Aldrich), and *tert*-butyl peroxide (TBPO, 98%, Sigma-Aldrich) were used as reagents without further purification.

Deposition of PDVB occurred at a chamber of 20 Pa, stage temperature of 13 °C, and filament temperature of approximately 350 °C. The flowrate of the TBPO was maintained at 5 sccm while the DVB vapor was allowed flow unmetereed from a container of liquid DVB at room temperature. The DVB feedstock was only 80% purity; the balance was composed of isomers of ethylvinylbenzene that incorporates in the polymer film as detailed previously [8]. Deposition of PGMA occurred at a chamber pressure of 26.5 Pa, stage temperature of 25 °C, and filament temperature of approximately 175 °C. The flowrate of the TBPO was maintained at 5 sccm while the GMA vapor was allowed to flow unmetereed from a container of liquid GMA heated to 40 °C.

2.2. Polymer spectral characterization

UV–vis transmission (Ocean Optics USB2000 + XR1 spectrometer with DH-2000-BAL deuterium/halogen light source) spectra were collected at normal incidence from ~1 μm samples deposited on a fused silica substrate. Fourier transform infrared (FTIR, Thermo-Fisher Nicolet iS10) spectra were collected on samples deposited on potassium bromide windows. Spectra were gathered in transmission mode and averaged over 32 or 64 scans with 4 cm⁻¹ resolution. In situ FTIR photo-oxidation experiments were conducted as described previously [4].

2.3. Film stress measurements

Film stress was measured by collecting 2-D interferograms (Zygo Verifire AT +) of 50-mm diameter silicon wafers coated with iCVD polymer. Wafers were double-side polished and interferograms were gathered on the uncoated surface to eliminate thin-film interference of the deposited polymer. Interferograms were collected at room temperature (~22 °C) before and after coating. The change in the radius of curvature was computed by subtracting the interferogram before coating from the interferogram after coating and fitting the resulting image to a sphere using custom-built software programmed in MATLAB™ (R2013B, Mathworks). Radius of curvature was then converted to film stress using the Stoney equation [9]. The Stoney equation also requires knowledge of the average film and substrate thicknesses. The average film thickness on the wafer was determined by spectroscopic ellipsometry (J.A. Woollam, M-2000XI) on $n = 56$ different points. Thickness variation over a 50-mm diameter wafer was typically within 10% (1-σ). Wafer thickness was measured with a digital linear encoder (Sony Magnescale LH-10). Substrate material properties used in the Stoney equation were Young's modulus $E = 150$ GPa and Poisson's ratio $\nu_s = 0.17$.

2.4. Bonding procedure

Various metal foils (Goodfellow, 5–50 μm thick depending on the foil type) were coated with iCVD PGMA and bonded by pressing together between two glass slides while applying pressure with commercial binder clips. Assemblies were heated to 150 °C for 30 min under N₂ and allowed to cool to room temperature. In the case of bonding lead to vanadium, assemblies were loaded to 3000 lbs in a commercial press and held in this configuration during thermal curing and cooldown.

2.5. Imaging techniques

PDVB films on silicon substrates were fractured and the cross-section imaged via scanning electron microscopy (SEM, Hitachi SU8000). A separate SEM equipped with a focused ion beam (FIB, FEI Nova600i NanoLab with gallium ion beam) was used to ion mill (30 kV, 20 nA) a large sub-surface cross-section for imaging PGMA bonded foils. Curvature of bonded assemblies was assessed using a 3D non-contact optical profiler (ZYGO, NewView 8300).

3. Results and discussion

3.1. Ablators for ICF

Plasma polymers are known to photo-oxidize under visible light illumination [4]. This photo-instability is believed to be due to light absorbing defects caused by plasma-induced fragmentation of precursors during deposition [4,10,11]. Because iCVD is a plasma-free process, only the thermolytically labile peroxide initiator undergoes fragmentation [12]. The resulting radicals initiate free-radical polymerization without appreciably damaging the chemical structure of the monomer. We hypothesized that the lack of monomer fragmentation in iCVD would eliminate the defects responsible for light absorption and photo-instability of plasma polymers. Fig. 1a compares the UV–vis transmission of plasma polymer with iCVD PDVB. The plasma polymer has a broad absorption ranging far into visible wavelengths. The iCVD PDVB is transparent throughout the visible regime and displays the sharp transmission cutoff in the UV that is characteristic of an amorphous hydrocarbon polymer.

Because the absorption of light is the first step in photo-oxidation, the more transparent iCVD PDVB also exhibits a much higher resistance to photo-oxidation than the plasma polymer. We simultaneously gathered FTIR spectra while illuminating samples of PDVB or plasma polymer with 455 nm light. As these polymers photo-oxidize, a broad stretch corresponding to carboxylic acid appears at ~3200 cm⁻¹; the integrated area under this peak is indicative of the extent of photo-oxidation [13,14]. Fig. 1b compares the photo-oxidation of PDVB with plasma polymer. The plasma polymer photo-oxidizes rapidly under illumination, whereas the iCVD PDVB shows no signs of photo-oxidation. The transparency and photo-stability of PDVB relative to plasma polymer suggests that monomer fragmentation and subsequent defect formation does not play a large role in iCVD.

While the enhanced photo-stability makes PDVB an attractive material, layers must be grown up to 200 μm thick in order to fabricate ablator shells. The CVD growth of thick films is limited by the generation of intrinsic film stress. This stress results in increasing strain energy as films grow thicker; this stored energy may eventually manifest itself as film cracking, delamination from substrate, or substrate damage. While film stress is typically attributed to crystalline effects (e.g., grain boundaries or epitaxial mismatch) [15], even amorphous CVD materials exhibit film stress due to instantaneous surface stress during deposition [16]. For example, amorphous hydrocarbon plasma polymers deposit with 10–100 MPa compressive stress [17].

PDVB represents an excellent case study in film stress because it is a stiff, glassy, and chemically crosslinked material; therefore, stress

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