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Fabrication and characterization of thin polymer targets for laser-driven ion acceleration

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Abstract. Polymer foils with thicknesses 150 nm to around 1 μm were manufactured using the process of spincoating. Varying the concentration of the polymer solution was found to be the most efficient way to control resulting foil thickness. Both interferometric microscopy and energy loss of alpha particles were used for characterization and found to yield agreeing results. For an experiment on laser-driven ion acceleration, these films were used as targets and as substrates for cryogenic hydrogen targets.

1. Introduction

High power laser systems allow for investigation of laser-plasma-interactions and laser-driven ion acceleration for which several mechanisms have been presented in the past. While *Target Normal Sheath Acceleration* (TNSA) [1] takes place in an overdense and thus for the laser impenetrable plasma, laser intensities around $10^{21}\text{W}/\text{cm}^2$ enable relativistic transparency for which the ion acceleration mechanism *Laser Breakout Afterburner* (BOA) was presented [2]. This new acceleration mechanism does impose new requirements on both the laser pulse and target characteristics.

In the scope of this work, thin hydrocarbon foil targets with thicknesses in the range of about 150 to 1000 nanometers were manufactured. Equally important to the production process, a reliable method of characterization needed to be found to be able to analyze produced target foils and improve the fabrication process.

2. Choice of material

Due to their uniquely high charge-to-mass-ratio, hydrogen ions, consisting of a single proton, are ideal for acceleration by electromagnetic fields. However, pure hydrogen exists as a gas at room temperature and needs to be cooled below 14 K to form a solid target for laser-driven ion acceleration. Avoiding the inconvenience of cooling to this extremely low temperature, solid hydrocarbons provide proton sources that are easier to handle. Polymethylpentene was chosen for this work for its high hydrogen content with a carbon to hydrogen ratio of 1:2.



3. Manufacturing

Thin foils of solvable materials can be produced using the process of spin coating where the solution is placed on a substrate on a rotatable disk. While the disk is spinned (hence the name of the process), the solution spreads out due to centrifugal force, the solvent evaporates and a thin film of solid remains.

3.1. Preparations

Polymethylpentene is available as a granulate, solvable in cyclohexane at 60°C, which takes about one hour. Preparation of substrates involves application of a separation layer on the substrate as a soluble and thus removable base. This enables foil removal from substrate, to get freestanding targets. Water is a solvent that is easy to handle and doesn't affect the polymer foils, so tensides were used as a water-soluble separation layer.

3.2. Choice of parameters

We found that the resulting film thickness depends on flow velocity during the spin coating process which can be manipulated by different means. Rotation speed of the spinning disk determines centrifugal force while a change in concentration of the polymer solution changes its viscosity, both affect the flow velocity and thus can be used to influence resulting foil thicknesses. As shown in figure 1, doubling the concentration of the polymer solution more than doubles the resulting film thickness, while doubling the rotation speed only decreases the resulting thickness by a small fraction. Therefore, the concentration was used subsequently as means to control the foil thickness.

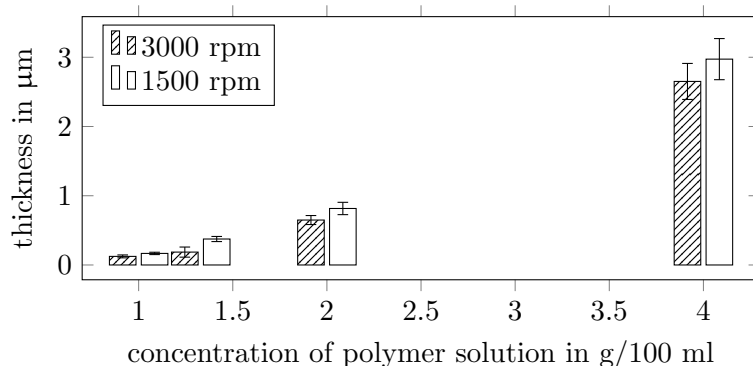


Figure 1. While doubling the rotation speed only decreases foil thicknesses by a small amount, doubling the concentration of the polymer solution more than doubles the resulting foil thickness. The latter has a much bigger influence and is consequently used to control foil thickness.

3.3. Removal from substrate

Target foils are removed from their substrate via dissolution of the separation layer. To achieve this, the substrate is placed in a container at an angle of ca. 45° relative to the horizontal (see figure 2). Then, the water level is raised continuously, thus dissolving the (water-soluble) separation layer and lifting the foil. Now it floats on the surface and can be lifted off. A useful tool for that is a small metal sheet with a hole in it. This creates a target foil with free front and back while the metal part can be glued onto a holder with a geometry tailored to fit the setup for the acceleration experiment.

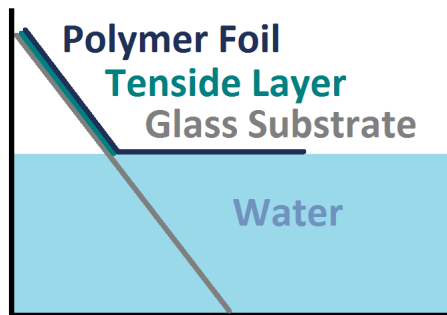


Figure 2. Setup for removal of foils from substrate, together with the order of layers on the glass substrate. A slowly rising water level dissolves the separation layer (middle) and lifts the target foil (top, dark) off its substrate (bottom, grey).

4. Characterization

Being able to measure the thickness and surface characteristics of produced foils is crucial to evaluate their suitability for experiments and adjust and optimize the production process. Two methods were used in the target laboratory of TU Darmstadt: Interferometric microscopy and energy loss of alpha particles. While interferometric measurements characterize the surface, alpha particles traverse the whole volume.

4.1. Interferometric thickness measurements

In our target laboratory, interferometric measurements can be carried out using an optical profilometer. This can be used for quantization of surface roughness and, via measuring the step height between foil surface and substrate surface at the edge of a sample, for thickness measurements. The latter can only be performed on witness samples, not on a foil that was already removed from its substrate and prepared for use in an experiment. Once a foil has been placed on a target holder, it doesn't necessarily lie completely flat anymore which might lead to a step height that is not identical with the thickness anymore. Another drawback of the procedure is that it doesn't give any information on the foil's backside.

An example for a surface profile can be seen in figure 3, corresponding to the lineout presented in figure 4.

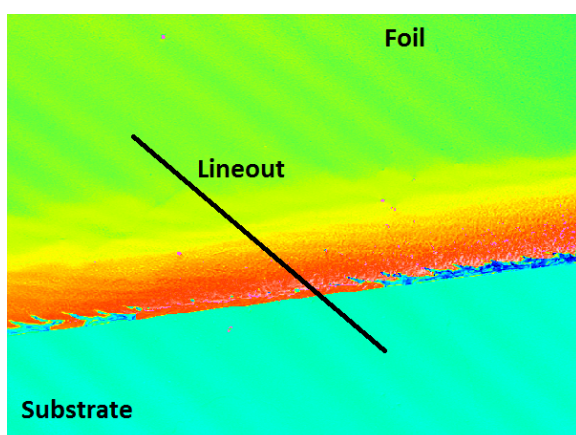


Figure 3. Interferometrically measured surface profile of a foil (upper half) on a substrate (lower half). It is visible that, from creating the edge and thus ripping the polymer foil off on the lower half of the picture, the edge has been disturbed and lifted from the substrate. Since the backside can't be measured, only the increase in height is visible. The black line marks the location of the lineout, displayed in figure 4.

4.2. Energy loss of alpha particles

Alpha particles lose energy while passing through matter mainly through interaction with electrons. With known probabilities for interaction events and energy loss per event, the total energy loss depends on the number of collision partners, thus translating (e.g. via simulation

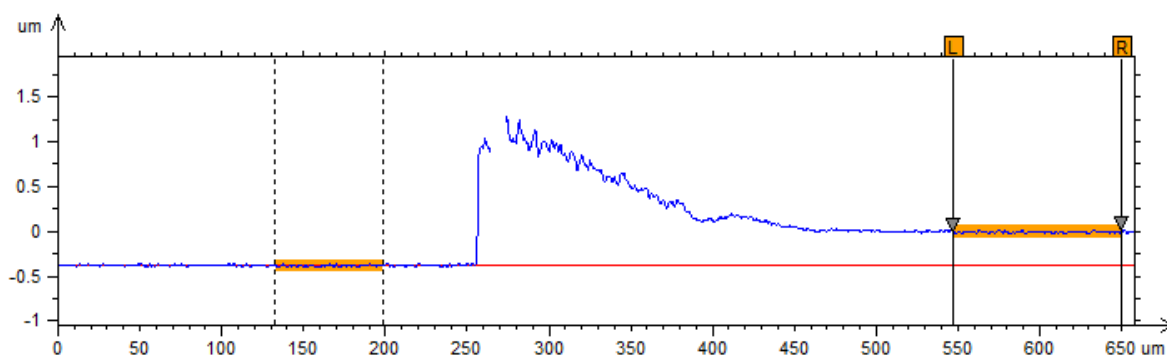


Figure 4. Lineout from the surface profile shown in figure 3. The artificially increased height from the lifted edge is clearly visible. The foil thickness can be determined via the step height, here taken as the height difference between the highlighted, unperturbed sections.

software) to the number of electrons the particle has interacted with. To convert this into thickness information, the material density is required which might be unknown or, in a thin film, different from the density of the bulk material.

Americium-241 was used as a source for alpha particles, it emits them with energies of more than 5 MeV. The energy spectrum is displayed in figure 5, together with the shifted spectrum after passing the sample. The total energy loss of the alpha particles was calculated using the energy shift of the main peak. Measurements of alpha particle energies were conducted using a Schottky diode consisting of gold and silicon. The particles lose their kinetic energy in the detector which creates electron-hole-pairs in the space-charge region. The total charge created corresponds to the amount of energy that was deposited.

A clear spectrum with sharp peaks is needed to be able to measure small energy shifts of the main peak. This requires a thin, thinly coated or uncoated and uncovered radioactive source, as the spectrum shifts and broadens as the alpha particles pass through the source material or coating.

For this work, conversions to thickness were carried out using simulations with the program SRIM (*Stopping and Range of Ions in Matter* [3]) using the density of the bulk polymer given by the manufacturer.

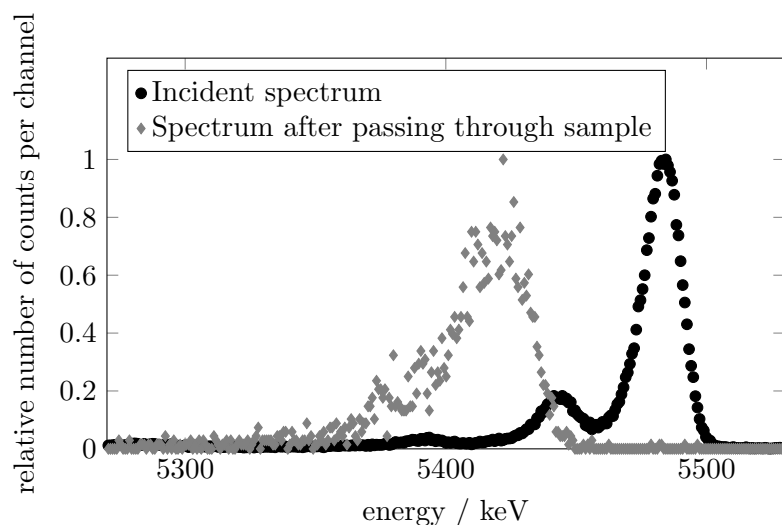


Figure 5. Energy spectrum of alpha particles emitted from Americium-241 (dots) and the shifted energy spectrum (diamonds) after the alpha particles passed through a polymer foil. The energy shift of 63 keV corresponds here to a thickness of 790 nm.

Compared to step height measurements, this method can characterize the exact same sample that is later used for the experiment. However, the measurement yields an average over the whole aperture since the alpha particles are not focused and the source size here was larger than the target size. Consequently, there is neither space-resolved thickness data nor surface information.

The method of thickness measurements via energy loss is promising for characterization of cryogenic targets and has already been utilized for this particular application before. However, due to melting and sublimation, cryogenic targets are not stable and therefore measurements are time-critical. To enhance time resolution, count rates must be increased. Regarding this, the setup can be improved via optimizing source activity, geometry of setup, aperture size and alignment. Considering geometry of setup, special care must be taken to ensure that a large number of alpha particles hits the detector after having traversed the target, while none are detected that didn't pass the sample. While a small degree of spatial resolution is possible with an array of small sources [4] or a mask effectively turning one bigger source into several smaller ones, this severely limits count rates and thus might not be applicable to all setups depending on free aperture.

We propose to describe target foils (of any material) by amount of energy loss rather than thickness. As the total energy loss relates to the number of electrons in the sample, such a description is connected more closely to the actual interaction mechanism between laser and matter which happens via electrons [1, 2]. In addition, the density of the material is not needed for characterization, which might not be provided to sufficient accuracy, depend on properties like chain length or branching in polymers [5] or is only known for the bulk material without insight in how much it might change during manufacture or treatment (coating techniques, sputtering etc. [6]). However, due to the geometry of the characterization process with alpha particles, description via total energy loss is in most cases only suitable for foils with uniform thickness across the aperture.

4.3. Comparison of methods

Results from both methods presented here are displayed in figure 6. Their results for foil thickness are in good agreement. In addition, it is revealed that resulting foil thickness is proportional to the concentration of the polymer solution used for the spincoating process (in the range of 1 to 2.8 g/(100 ml)) which makes thicknesses of new targets predictable.

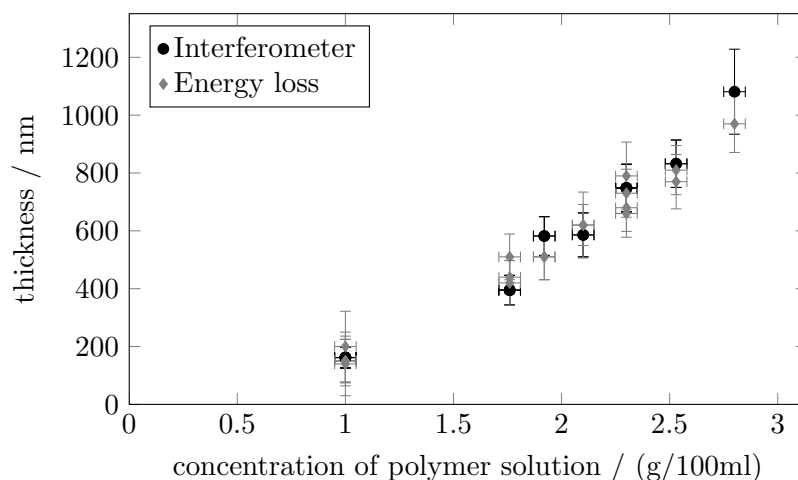


Figure 6. Comparison of interferometrically measured values for target thickness to those calculated from energy loss of alpha particles. A clear proportionality to concentration of the solution is visible, while the thickness measurements from both methods are in good agreement.

5. Experiment

In April 2013, thin foils of polymethylpentene were used as targets in experiments on laser-driven ion acceleration at the *Petawatt Laser for Heavy Ion Experiments* (PHELIX) at *GSI Helmholtzzentrum für Schwerionenforschung* in Darmstadt. The experimental setup included a tilted target to allow for an incident angle of laser direction on target (see figure 7). Some beam profiles exhibit two beam directions as can be seen on a stack of radiochromic film which is colored by ions (figure 8): One beam along target normal which is the expected direction for the acceleration mechanism of TNSA and one along direction of laser propagation which is the suspected ion beam direction for the BOA mechanism. These results indicate that the fabricated target foils were suitable for ion acceleration by not only the TNSA mechanism but also the BOA acceleration scheme (see also [7]).

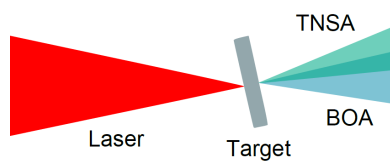


Figure 7. Geometry of experimental setup with expected ion beam directions. The target normal is tilted with respect to laser direction.

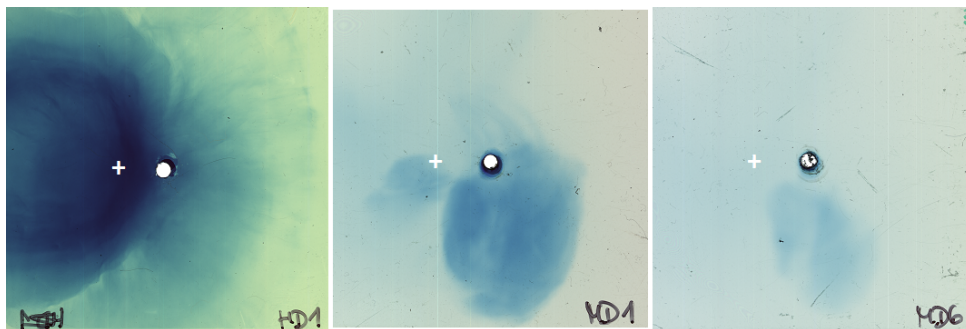


Figure 8. Ion beam profile on radiochromic film, corresponding to ion energies from lower towards higher energies (left to right), exhibiting two distinct beam directions with different maximum energies; one along the target normal (white cross) and one in laser direction (hole in center).

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