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BACKLIGHTER TARGETS**

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FABRICATION AND CHARACTERIZATION OF ALUMINUM OXIDE AEROGEL BACKLIGHTER TARGETS

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Aluminum oxide aerogel can be used as a backlighter target to provide a radiation source for diagnostics during ICF experiments. To demonstrate the feasibility of this type of target, it was necessary to cast thin pieces of aerogel for plasma emission studies of aluminum oxide. We were able to demonstrate density control over a range of 50–400 mg/cc, and, furthermore, cast the aerogel as a thin (0.6–0.7 mm), smooth monolith that did not require additional machining. The fabrication of these targets begins with a solution of aluminum chloride, ethanol, and water and is then catalyzed with propylene oxide to gel within molds to form the shape. Supercritical drying with carbon dioxide provides the dry aerogel. Various target densities were made by adjusting the relative amounts of starting materials and post treatment condition. The finished materials were characterized for density, pore size, and water content. Initial freestanding targets of 98 mg/cc have been fabricated and shot, and other similar targets of densities from 50–400 mg/cc have been fabricated for future experiments.

I. INTRODUCTION

Aluminum is one of several materials of interest as a backlighter for ICF and related high energy density physics experiments.¹ Typically the target consists of a mass limited, full density foil.² In this work, low density (50–400 mg/cc) aluminum oxide aerogel targets were fabricated. These targets are being used for an ongoing series of short-pulse backlighter experiments by researchers at the University of Rochester to study the effect of target density on emission intensity.³ The specifications for this target are for a thin (0.6–0.7 mm) freestanding aerogel with controlled density. In order to assure reproducible plasma characteristics from shot to shot, it is important that the areal density of the material is very uniform. This requires the pores within the target be very small compared to the wavelength of the incident laser radiation, the thickness deviation across the target to be small, and the surface to be relatively smooth.

The classic method in literature for producing alumina aerogel is the Yoldas method^{4,5} in which aluminum sec-butoxide is used as the precursor. Recently epoxides have been used as catalysts for sol gel reactions of inorganic salts⁶ and were chosen as the synthesis method for these targets. This synthesis route is less moisture sensitive before gelation and allows a more easily reproducible procedure. In this work we expand this method to produce a wide range of densities to create a different type of backlighter than is typically used.

II. EXPERIMENTAL

Freestanding, flat sheets 0.6 to 0.7 mm thick and 1 to 9 mm² in area were fabricated as shown in Fig. 1. These aerogels were cast from a 50% vol/vol solution of water and ethanol (absolute, Aldrich) with aluminum chloride hexahydrate (AlCl₃·6H₂O, 99%, Aldrich). An excess of propylene oxide (99%, Aldrich) equivalent to the volume of ethanol or water was then added to catalyze gelation. (All chemicals were used without additional purification or preparation.) A series of different concentrations of AlCl₃·6H₂O between 0.069 M and 1.1 M were prepared in order to measure the effect of density. To cast thin flat pieces, the solution was subsequently poured into a container with molds made from quartz slides as shown in Fig. 2. The solution was then allowed to gel under dry N₂



Figure 1. Picture of a 98 mg/cc alumina aerogel of 0.7 mm thickness.

for 24 h. Ethanol was then added on top of the solid gel and allowed to cure for 24 h. The targets were cut to the correct lateral dimensions after removal from the mold, prior to drying. The gels were then solvent exchanged into isopropanol (IPA) over a period of 5 days to remove the remaining water and unrelated precursor materials in the gels. The CO₂ drying process consisted of placing the hydrogels in a pressure vessel and flushing with liquid CO₂ for 5 days, then heating the CO₂ and venting from supercritical conditions (1200 psi and 35°C). There was substantial shrinkage of the gels during the solvent exchange to IPA and CO₂ drying steps. The gels would lose about one third of their thickness, so that wet gels were cast to 1 mm thickness to achieve the 0.6 to 0.7 mm specified thickness.

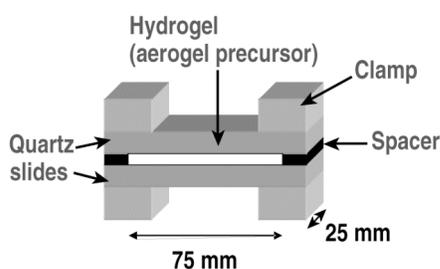


Figure 2. Diagram of mold used in casting the 0.7 mm thick alumina aerogel targets.

Although these samples were dried with the supercritical CO₂ drying process, trial drying runs were done in supercritical isopropanol (IPA) as well. In these trial runs the aerogel was solvent exchanged into IPA and sealed in a pressure vessel with excess IPA. The temperature was then ramped at 1°C/min to 270°C, and left to dwell at this temperature for ~2 h. During this time the pressure increased to ~1200 psi. The supercritical IPA was then vented, providing a dry aerogel.

For density measurements, the volume was measured with a calibrated microscope and weighed with a microbalance in ambient lab conditions. Thermogravimetric measurements were completed with a TA instruments Q500 with nitrogen as a purge gas. Surface roughness data is from scanning white light interferometry of the samples with a Veeco NT3300. Due to the transparency of the pieces, it was necessary to sputter coat the sample with ~300 nm Au in order to get adequate reflection from the surface prior to the roughness measurement. [Although deposition of the gold layer could cause minor changes in the surface of the sample, the size of the defects of interest (>1 μm) are large enough that this coating does not significantly alter the surface.] Scanning electron microscopy (SEM) images were taken with a JEOL JSM-6460 after coating the samples with <100 nm Au. After degassing the sample under vacuum at 100°C

for 12 h, nitrogen adsorption was measured with a Micromeritics ASAP 2020 nitrogen adsorption analyzer. Brunauer-Emmett-Teller (BET) and Barrett-Joyner-Halenda (BJH) analysis^{7,8} of the isotherm data was performed with the ASAP 2020 v3.0 software.

III. RESULTS AND DISCUSSION

By casting and drying a series of cylindrical samples it was found that the density could be controlled from 25-200 mg/cc by adjusting the aluminum chloride concentration. At the lower limit, the density was limited by the inability of the solution to fully solidify during the gelation below concentrations of ~0.1 moles/L (M). This led to gels that would not retain their shape after separating from the cylindrical mold. At a concentration of 0.069 M, the gel was soft and shrank more than the gels made with higher concentrations. At concentrations >0.069 M, there was roughly a linear relationship between density and amount of aluminum chloride added (Fig. 3). For concentrations higher than ~2 M a practical limit was reached due to the difficulty of dissolving all the aluminum chloride in the starting solution.

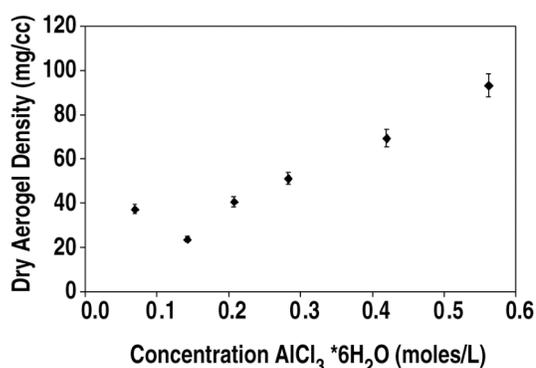


Figure 3. Plot of starting concentration of aluminum chloride in precursor solution and density of the final dry aerogel.

It was not surprising that a lower limit was reached in which the solution would not fully gel. What was unexpected was the extra shrinkage of the gels made with 0.069M AlCl₃·H₂O (Fig. 3). This is likely due to syneresis of the gels, in which the alumina network contracts and expels the pore liquid.⁹ Qualitatively, dramatic shrinkage occurred to all the alumina gels we fabricated if they were never dried and left in IPA over a period of months. This suggests that the issue is one of different rates of shrinkage between the lowest concentration gel and the others. It is known that larger gels (with a smaller surface to volume ratio) undergo this process more slowly than similar small gels because of the liquid escapes more easily from the gel.⁹ Because of this, it is likely that the higher concentration samples with a more densely packed

network likewise inhibit the loss of liquid. If this is true, the shrinkage for the lowest concentration can be limited in the future by decreasing the IPA exchange time or using a different solvent. (It is important to balance this with removal of propylene oxide because the gel can re-dissolve in this solvent over time.) If the goal is to increase the density, however, this may provide another method of control.

It is advantageous also to be able to cast these targets to the required dimensions in order to avoid machining, which is especially time consuming for aerogels. For the thin (0.6 mm) cast targets, the lowest achievable density was limited to 50 mg/cc or greater. This is because of the stress put on the gel during separation of the wet gel from the mold. For the cylindrical test pieces, the gels were compressively stressed as they were pushed out of the mold. The flat pieces were removed from their molds by sliding and prying the mold apart, causing tensile stress that ultimately caused the lowest density gels to tear down the center as opposed to cleanly separating.

Removal of the flat targets from the mold was most likely the cause of much of the surface roughness of the final target. A number of surface roughness measurements were made on representative, dry ~100 mg/cc density flats. The surface roughness varied greatly over different locations on the surface, with rms values from 150 nm to 875 nm over length scales of 200 x 300 μm . The surface roughness was dominated by large grooves and depressions (Fig. 4) while the background roughness of areas between defects was ~100 nm rms. The grooves seem to have the same preferred direction, and were likely cause when the material was separated from the quartz slides. Although these defects were present, the targets meet the specification of <1 μm rms roughness.

Although we also describe a supercritical IPA procedure for drying, CO_2 drying was chosen as the baseline process for these targets. This is because of optical non-uniformities in the supercritical IPA process. The CO_2 dried gels were very transparent, but those dried with the IPA process described above were not all optically uniform. Some parts of those gels were translucent to opaque indicating a difference in the pore or microstructure of the material. In the future, this process may be refined by changing the drying parameters.

The aerogels fabricated with the CO_2 drying process have a small pore structure as shown by SEM (Fig. 5). N_2 adsorption (Fig. 6) data confirms this, with a BJH average pore size of 11.5 nm. This single value represents a wide distribution, however, and a large number of pores exist from 1 to 100 nm. No pores greater than or equal to 1 micron were seen in any of the SEM images and this is confirmed by the optical transparency of the aerogels

fabricated. (Pores of this size or greater would render the aerogel cloudy or opaque.) Due to the high porosity and small pore size the surface area was very high, with a measured BET surface area of 100 mg/cc samples of 330 m^2/g .

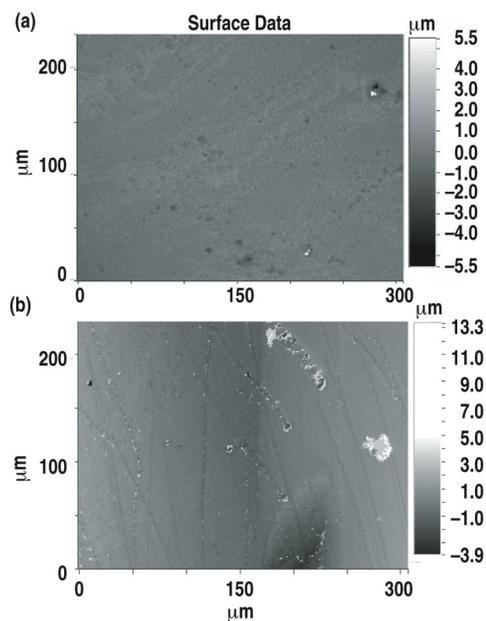


Figure 4. Contour plot of 93 mg/cc alumina aerogel surfaces from an optical profiler. Image (a) shows a representative area with a number of small depressions, while (b) shows the deep grooves that can be formed.

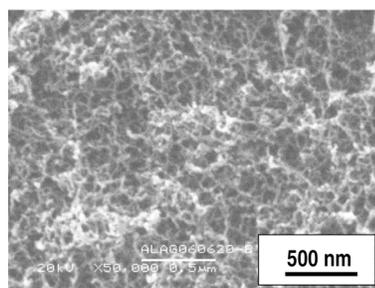


Figure 5. SEM of alumina aerogel.

The density can also be increased post process by heat-treating the dry aerogels (Fig. 7). By heating under dry N_2 , the alumina aerogel shrinks and increases in density while retaining its basic shape. During this process, water is also lost from the aerogel. As fabricated before heat treatment, the aerogel contains 39% water. At 800 $^{\circ}\text{C}$, all the water of the alumina (hydrate) aerogel is evolved from the sample (Fig. 8). A dramatic weight loss is seen at low temperatures corresponding to loss of loosely bound, adsorbed water. At higher temperatures, water that is chemically bonded is released. Understand-

ing both the shrinkage and water loss enables estimation of the elemental aluminum content which can then be verified via elemental analysis.

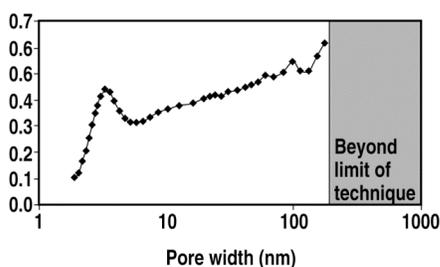


Figure 6. BJH distribution of alumina pore size calculated from desorption data of 100 mg/cc aerogel.

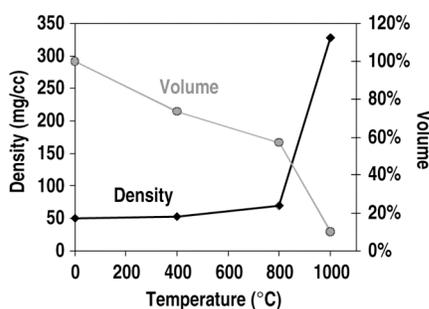


Figure 7. Plot showing that higher temperatures affect the density and volume of the aerogel. By heating the alumina aerogel, higher densities can be achieved and water is removed.

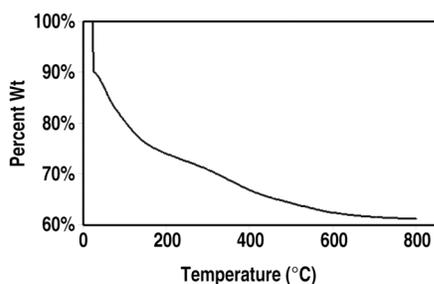


Figure 8. Plot of weight versus temperature from thermogravimetric analysis (TGA). A dramatic weight loss is seen at low temperatures corresponding to loss of loosely bound, adsorbed water. At higher temperatures, water that is chemically bonded is released.

Freestanding targets of 98 mg/cc density were fabricated as described above at General Atomics and shot at the Rutherford Appleton Laboratory.³ The incident laser had an intensity of 1.5×10^{18} W/cm², 0.7 ps pulse length, and approximately 100 μ m focal spot on the target. In the same series full density foil and porous

alumina were shot for comparison and the emission spectra of the targets were measured in the range of 6.2-8.0 Å. The preliminary data obtained from this initial experiment is promising and additional experiments will be performed at LLE.³

IV. CONCLUSION

Alumina aerogels were successfully fabricated and characterized at 0.6-0.7 mm thickness. We were able to produce a range of densities from 25-400 mg/cc in cylindrical castings, and 50-400 mg/cc densities in the thin flat targets. Future work will continue on expanding the range of densities, improving the surface finish, and investigating supercritical drying in other solvents besides CO₂.

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