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ABSTRACT

We have successfully sputter coated sub-micron layers of quartz onto plasma polymer shells. An agitation mechanism based on an electromagnetic shaker was used. Coatings as thin as 0.4 µm that retain their integrity have been deposited. These coatings have permeation rates against helium at room temperature that are similar to those of thermal quartz. However, the permeation rates to D_2 and argon of coatings thinner than ≈ 2 µm are higher than expected. In contrast, coatings thicker than 2 µm had D_2 half-lives that were long enough to make them useful as a permeation barrier. Diffusion along grain boundaries or through pinholes is a likely reason for the high permeation rates through the thinner coatings. Because plasma polymer becomes thermally unstable near 300°C, these composite shells have to be filled at a maximum temperature of 250°C.

I. INTRODUCTION

Several target designs for the National Ignition Facility (NIF) involve plastic mandrels[1] made by plasma polymerization and the depolymerizable mandrel technique.[2] Because the permeation rate of D_2 through a plasma polymer shell is very fast (half-life of less than one hour), these targets will require a permeation barrier if room temperature handling or experiments are required. Polyvinylalcohol (PVA) has been used successfully as the permeation barrier for Nova shells for many years.[3] However, the PVA coating process has a very low yield (≈ 3%) and the final coating is rather "lumpy," leading to undesired wall thickness variations around the shell.[4] Therefore, an alternative to this technique is highly desirable.

Glass has ideal permeation characteristic against D_2. D_2 can be permeated through a thin glass shell at elevated temperatures (≈ 360°C) in a few days, but will be trapped inside the glass shell at room temperature for months. Therefore, we explored the possibility of providing plasma polymer shells with an alternative permeation barrier by sputter coating them with glass. The presence of thick glass layers (> 1 µm) in ignition targets has been traditionally undesirable, although a current design does incorporate rather thick glass layers.[5] Therefore, we pursued determining the thinnest possible coating that would retain its integrity and provide good permeation characteristics against D_2. We initially determined helium permeation rates and also explored the possibility of permeating argon into the glass coated shells as well.

II. EXPERIMENTAL

Plasma polymer shells, ≈ 900 µm in diameter and 10 to 20 µm thick, were used as the starting mandrels. Sputtering was performed in a Temescal sputtering system. All sputtering runs were done at 500 W at 13.6 MHz. The system pressure was varied to obtain the densest coatings as described below. 99.95% pure quartz discs were used as the sputtering targets. A Labworks electromagnetic shaker driven at 10 to 1000 Hz was used as the agitation system. Shells were bounced in pans normally used for plasma polymer coatings mounted onto the electromagnetic shaker. The ideal driving frequency was about 60 Hz. At this frequency the mandrels skated around the bounce pan instead of bouncing up and down. The coated shells were examined optically and by scanning electron microscope (SEM) for microcracks and general integrity. The coating thickness was measured by interferometry. We measured the permeation half-life of helium, argon and D_2 at room temperature as a measure of the permeability of the coatings to these gases. The coated shells were first permeation filled with the desired gas at elevated temperatures and then the rate of outgassing of that gas was measured in several ways. Weight loss and diameter shrinkage of the filled shells as a function of time provided half-lives to helium and D_2. Direct outgassing into a sealed evacuated volume was also used for D_2 and helium. The decrease in the x-ray microfluorescence signal from argon inside the shell was used to determine half-lives to argon. Consistency of results obtained from the above methods provided added confidence in the measured half-lives.

III. RESULTS AND DISCUSSION

The background pressure in the coating process can affect the structure and quality of the coating tremendously. Figure 1 shows this for our sputtered quartz coatings on flat Si wafers. The coating deposited at 2 mTorr appears very dense in cross-sectional SEM examination, while the one deposited at 10 mTorr has a columnar structure. Therefore, we performed all our coatings at ≈ 2 mTorr. This was the lowest pressure at which we could reliably sustain a plasma in our system.

We also found out that the coatings develop large cracks, if the shells are placed too close to the sputtering gun during coating. The plasma polymer shells in such coating runs were found to have expanded. Although, a thermocouple gauge indicated that the bounce pan temperature was only 150°C, the shells must have experienced a higher effective temperature due to the flux of the sputtered material. In our system, the distance between the gun and the shells had to be at least 5 cm to avoid this problem. This led to a maximum coating rate of ≈ 0.3 µm/hr.

We also encountered unexpected problems when pressure filling the coated plasma polymer shells. Pressure filling of glass shells is usually performed at 360°C to expedite the filling process due to the increased permeation of fill gases through glass. Thermogravimetric analysis of plasma polymer had indicated that it should be thermally stable up to 400°C.[6] However, we found that prolonged exposure to 360°C in the filling process, led to dissociation of the inner plasma polymer mandrels. We eventually found that such fills had to be performed at 250°C or lower to avoid damage to the plasma polymer mandrel.

We initially attempted depositing as a thin a coating as possible. We found that the coating developed thousands of microcracks at a coating thickness of 0.2 µm (Fig. 2). This limited the coating thicknesses to 0.4 µm or greater. We deposited coatings as thick as 3 µm. The results of our experiments are summarized in Fig. 3. The helium permeation half-lives compare well to those of similar size thermally prepared glass shells. The half-life appears to have a linear dependence on the glass thickness. However, the situation is markedly different for D_2

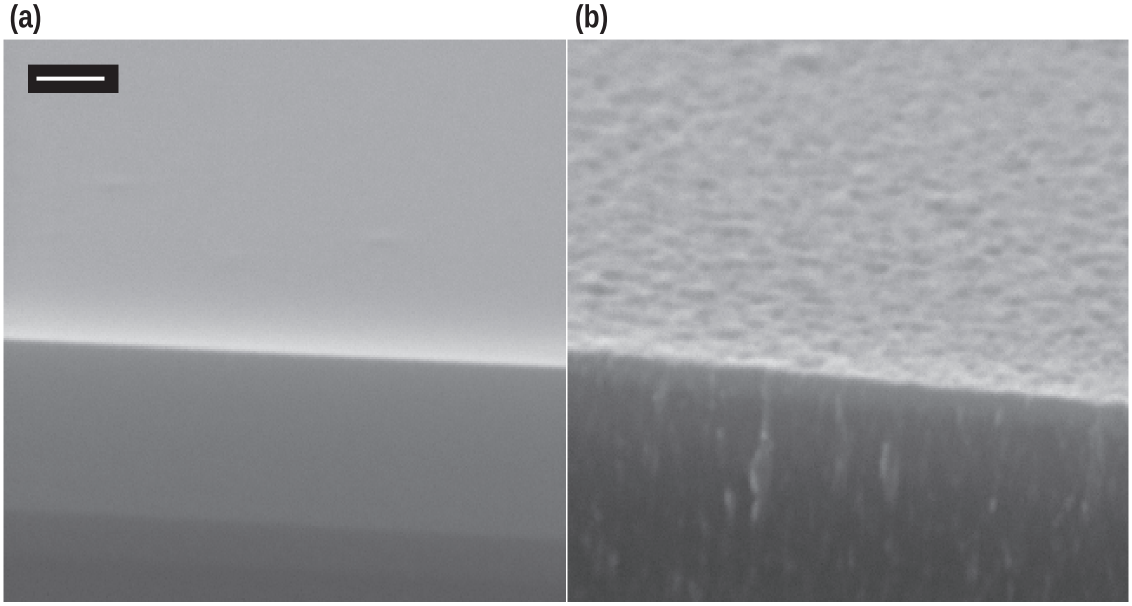

Fig. 1. SEM cross-sectional images of glass coatings deposited on a silicon wafer at (a) 2 mTorr, and (b) 10 mTorr, background pressure. The film in (a) appears very dense while columnar structure can be clearly seen in the film in (b). The scale bar is 3 µm.

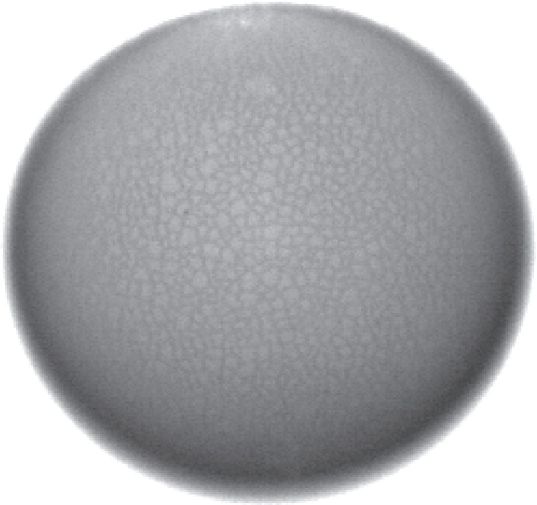

Fig. 2. Optical image of a 0.2 μm glass coating on a 900 μm diameter plasma polymer shell. Numerous microcarcks can be seen in the thin glass layer. This problem was not observed in thicker coatings.

and argon. The D_2 half-life is much lower than expected for coating thicknesses of less than ≈ 2 μm. For comparison, a 900 μm diameter, 0.5 μm thick thermally prepared shell has a D_2 half-life of many days. Thermally prepared glass shells are virtually impermeable to argon at room temperature or at 250°C, having half-lives of several years or longer. Our coatings, however, were permeable to argon. Shells with coatings of all thicknesses in the range we investigated could be filled with argon at 250°C. When the coating thickness reached ≈ 2 μm, the D_2 and argon half-lives increased dramatically compared to thinner coatings. At room temperature, coatings thicker than ≈ 2 μm had much longer permeation half-lives when compared to thinner coatings. These results were puzzling when compared to results for helium.

Closer examination of the sputtered glass coatings on shells revealed a very granular structure at the sub-micron scale. Figure 4, shows SEM and atomic force microscope (AFM) images of a 2.2 μm coating on a plasma polymer shell. The fine granular structure is evident in the AFM image. In addition, a pinhole type defect can be seen in the AFM image and similar defects can be seen in the SEM image. The SEM image also shows the presence of larger nodules scattered about the surface. Pinholes may have resulted when large nodules were knocked off the surface when shells collided against each other during bounce coating. The presence of numerous grain boundaries and pinholes in the coatings on shells makes the observed relatively fast permeation rates for argon and D_2 somewhat understandable. For thinner coatings pinholes could be a dominant escape route for argon and D_2 [Fig. 5(a)]. In thicker coatings pinholes may be covered up and diffusion can only take place along grain boundaries and through the bulk. In addition, grain displacement in thicker coatings will present a tortuous path of diffusion that further slow permeation of argon and D_2. On the other hand, helium permeation through the bulk is fast enough that the molecules may diffuse through the bulk and grain boundaries equally rapidly (Fig. 5(b)]. Therefore, the observed helium permeation rates are essentially the same as bulk diffusion and compare closely to thermally prepared glass shells. Microcracking may also be responsible for the observed diffusion rates, however, we could not find any evidence of microcracks even upon very careful examination of the coatings. In addition, the reproducibility of the data for any given coating thickness also makes the microcrack argument less likely.

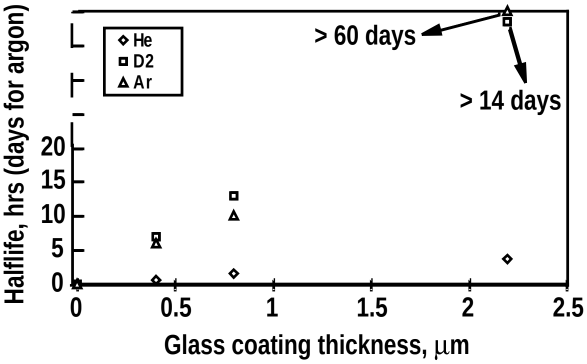

Fig. 3. Results of the permeation half-life measurements of argon, He, and D_2 through glass coatings of various thicknesses on 900 μm diameter plasma polymer shells. Zero coating thickness corresponds to values for bare plasma polymer shells. Note the different scale on the y-axis for argon. The values for He are similar to what is observed for thermally prepared glass shells of similar dimensions. However, permeation half-lives for D_2 and argon for coatings thinner than ≈ 2 μm are appreciably lower than those expected for thermally prepared shells (see text). The permeation half-lives increase dramatically for coating thickness of ≈ 2 μm or greater.

IV. CONCLUSION

We have been able to bounce coat plasma polymer shells with sputtered glass. 0.4 μm of coating thickness is required to obtain coatings that retain their integrity. The presence of pinholes left over by leaving nodules and the granular structure of coatings on shells leads to fast D_2 and argon diffusion rates for coatings thinner than ≈ 2 μm.

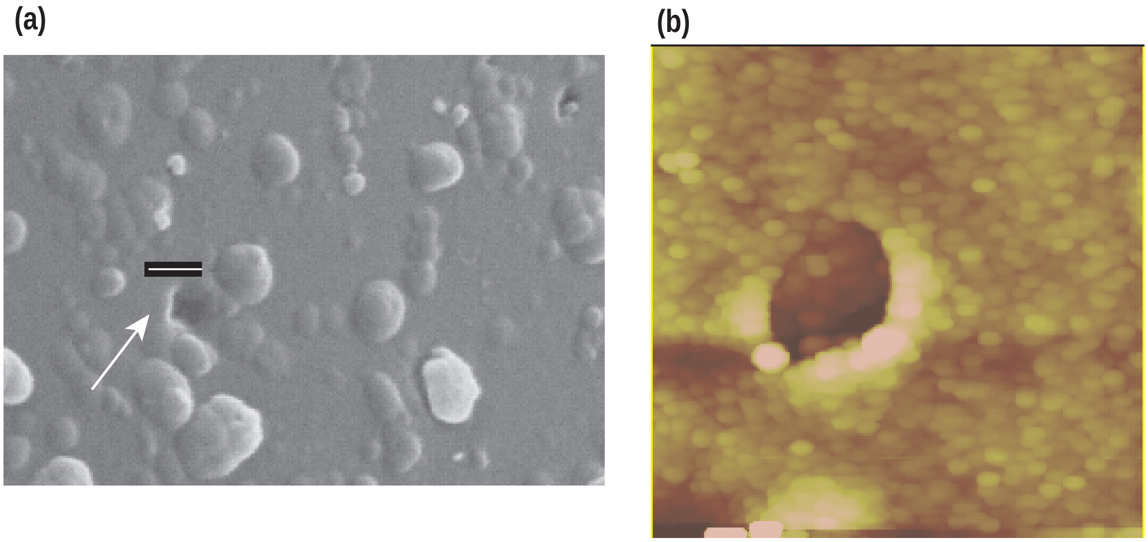

Fig. 4. (a) SEM image of a typical 2 µm coating. Notice the pinholes type defects present in the surface (white arrow). The scale bar is 1 µm. (b) 5 µm square AFM image of the same type of coating reveals a granular sub-micron structure. The scan was done around a pinhole. Notice for the thicker 2 µm coating, there is a glass underlayer inside the pinhole.

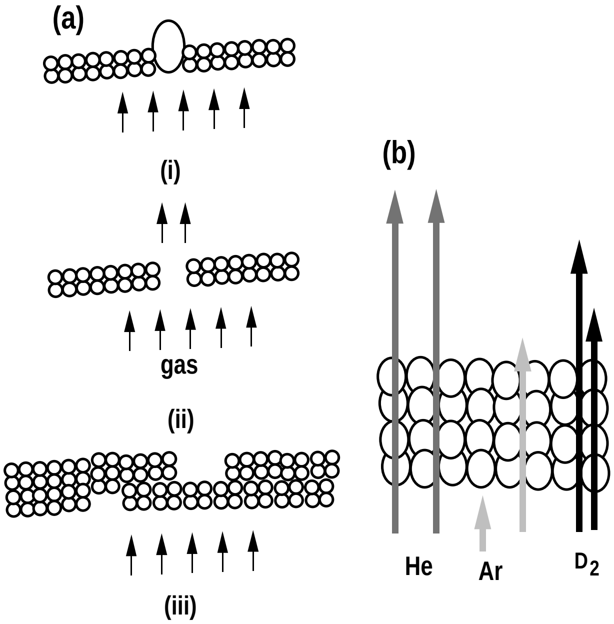

Fig. 5. Possible explanation for the observed rapid permeation of D_2 and argon through thin glass coatings. (a) Pinholes left behind (ii) by leaving large nodules (i) allow rapid escape of D_2 and argon. The pinholes are covered (iii) in thicker coatings. (b) He permeation half-life is not affected by the glass coating structure can diffuse through the bulk as fast it can escape along grain boundaries. D_2 and argon diffuse through grain boundaries much faster than through the bulk.

Coatings thicker than ≈ 2 µm can be used as an effective permeation barrier against D_2, while allowing the incorporation of diagnostic gases such as argon.

ACKNOWLEDGMENTS

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