Sputter-Deposited Be Ablators for NIF Target Capsules

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"SPUTTER-DEPOSITED BE ABLATORS FOR NIF TARGET CAPSULES"

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ABSTRACT

We have performed a series of preliminary experiments to determine whether sputter deposition of doped Be is a practical route to producing NIF target capsules with Be ablators. Films ranging in thickness from 7 to ~ 120 μm have been deposited on spherical polymer mandrels using a bounce pan to ensure uniform coating. With no voltage bias applied to the pan, relatively porous coatings were formed that were highly permeable to hydrogen. The surface finish of these films ranged from ~ 250 nm rms for 13- μm -thick films to a minimum of ~ 75 nm rms for an 80- μm -thick film. Application of a voltage bias was found to significantly modify the film morphology. At a bias of 120 V, 7- μm -thick films with a dense, fine-grained microstructure were produced. These capsules had a reflective surface with a 50 nm rms roughness. Finally, to demonstrate the ability to produce a graded dopant profile, a coating was produced in which the concentration of added Cu was varied from 2.5 atom % at the beginning to zero after 40 μm of deposition.

INTRODUCTION

The National Ignition Facility (NIF) will require spherical shell targets about 2 mm in diameter with a 120 to 150- μm -thick ablator.^[1,2] There are two basic capsule designs. One calls for a Ge-doped CH ablator while the other requires a Cu-doped Be ablator. In both cases, the doping level ranges from less than 1 to a few atom %, and the rms surface finish should be on the order of 10 nm or less. For indirect drive ignition experiments both designs call for a smooth, 80 to 100- μm -thick DT ice layer on the capsule's inner surface.

For the same outer surface finish, the Be design offers performance advantages over the CH design. Because of the lower atomic mass of Be, it has improved

hydrodynamic stability during an implosion. Calculations show that a Be capsule can tolerate increased levels of both short-wavelength roughness on the outer surface and roughness of the cryogenic layer before its performance degrades. In addition, simulations indicate that at reduced drive temperatures, Be capsules perform significantly better than CH. Because of these advantages plus the desirability of having as many target options available as possible, an effort is underway at LLNL to develop a target incorporating a Be ablator.

Any technology used for creating a Be ablator target must meet certain minimum requirements. First, it must be possible to uniformly dope the Be with up to a few atom % of a suitable high-Z material to enhance x-ray absorption. Copper is an acceptable material and in addition is soluble in Be at this level. Second, the technique used must produce a wall with a thickness uniformity of better than 1 μm and a surface roughness better than about 10 nm rms. Finally, the resulting capsule must be capable of being filled with and holding the DT fuel.

Beyond these minimum requirements, there are a number of other desirable characteristics. Most important among these is that the capsule have sufficient strength so that after filling with the required amount of DT, it can withstand the pressure (up to 350 atmospheres) at room temperature. This will allow room temperature handling of the capsule until it is positioned at the center of the target chamber and cooled to form the cryogenic layer. Eliminating the need for cryogenic handling from the time of fill adds flexibility to our fielding options and represents a significant cost savings.

The strength of a capsule prepared by any technology will ultimately need to be tested experimentally, as the processing history can greatly affect material properties. However, to gain a sense of the

tensile strength of Be. By applying the hoop stress equation

$$P = \frac{2t\sigma}{r},$$

where P is the pressure at which the capsule yields, t is the wall thickness, σ is the yield strength of the wall material, and r is the capsule radius, we can determine whether the strength required might be attainable with Be of the design thickness. Figure 1 shows the approximate range of reported values^[3] for yield strength (the point at which the material undergoes a small, typically 0.2%, plastic deformation) along with the pressures required for two different Be capsule designs. The strength values cover a broad range, due mainly to differences in fabrication techniques and material purity. It must be noted that none of the data found were directly applicable to the case of sputter-deposited films. The conclusion to be drawn from this plot is that it might be possible to produce beryllium coatings that are strong enough to hold fill at room temperature, but given the significant uncertainties in what strength can be achieved, the safety margin is quite small.

Fig. 1. A plot showing the yield pressure as a function of material strength for a 2-mm-diam capsule with either a 120 μm or 150 μm wall thickness. The black dots indicate the required room temperature fill pressure for each design. The shaded region reflects the range of values quoted in the literature for the yield strength of Be.

There are three approaches being pursued as routes to a Be ablator capsule. The most direct approach is to machine two hemishells from bulk Be and bond them together. Among the issues that will need to be addressed are the strength of the joint and its affect on the capsule performance, a suitable means to fill the capsule (since it will almost certainly be impermeable), and whether feedstock can be obtained with the required homogeneity and concentration of dopants. Another approach involves the use of Chemical Vapor Deposition (CVD) with an appropriate Be-containing compound. Issues particular to this process include finding a mandrel capable of withstanding the elevated temperatures required, and eliminating contaminants in the coating that originate from the Be monomer. CVD films are typically dense, and therefore impermeable, so as in the case of machining a suitable drill and plug technology would be necessary to fill the capsule.

At LLNL we have been exploring the third option of sputter-deposited Be coatings as a route to NIF ablaters. As with CVD, this approach requires the use of a mandrel on which the Be is deposited. It allows considerable flexibility in adding dopants, including the capability of providing a non-uniform dopant profile. We have demonstrated that NIF-scale thicknesses of Cu-doped Be can be applied to plastic mandrels using sputter deposition. The next section provides some experimental details, followed by our results to date. Finally, we discuss the remaining key issues and our plans to address them.

APPARATUS

The system used for Be deposition on capsules is a conventional stainless steel bell jar equipped with a cryopump, which provides base pressures in the 10⁻⁸ torr range. Three unbalanced planar DC magnetrons with 3.3-cm-diam targets are fixed to the top of the chamber. For most of the experiments described here, two of the targets were pure Be, and the third was Be/6 atom % Cu. The magnetrons point down towards a 5-cm-diam concave pan that holds the samples. This pan is in turn mounted on a shaft that is bonded to a piezoelectric crystal, which is excited by a 35.2 kHz pulse train during the deposition. The resulting motion of the pan causes the capsules to bounce and roll randomly while the Be is depositing, producing a uniform coating. The bounce pan is electrically isolated, so if desired a voltage bias can be applied. In addition to providing argon for the sputter

guns, the gas handling system includes a high-speed valve which allows pulsed dosing of a reactive gas during deposition. The magnetrons have been operated at up to 175 watts each, but are normally run at 125 W. The power to each gun can be adjusted independently to vary the doping level. Figure 2 shows a schematic view of the apparatus.

Fig. 2. A schematic diagram of the sputter deposition chamber.

CHARACTERIZATION

The main emphasis of the characterization effort was to determine the grain structure and roughness of the films. The principal tools were scanning electron microscopy (SEM) and atomic force microscopy (AFM). SEM images were obtained from outer surfaces as well as fracture cross sections. AFM images, from which roughness values were calculated, were restricted to 5 to 20 μm square scans on the outer surface. Therefore, the quoted roughness values exclude perturbations at longer wavelengths. In addition, the capsules were weighed to gauge the density of the films, and a limited amount of x-ray fluorescence (XRF) analysis was performed to monitor contamination and dopant concentration.

RESULTS

Most of the data shown here were obtained by depositing Be on thin-walled polymer capsules. The procedure used to fabricate these mandrels has been described in detail elsewhere.^[4,5] Typical Be deposition runs last for 7 to 8 hours. Depending on the deposition conditions, this yields a film from 7 to 14 μm thick. Although these films are much thinner than those required for NIF target capsules, they are thick enough to establish a microstructure that changes little with additional thickness. Extended runs have been performed to verify this behavior, yielding data on films as thick as $\sim 120 \mu\text{m}$.

Figure 3 shows a cross section SEM image of a 13.4 μm thick Be film deposited on a 3- μm -thick capsule. The pronounced columnar structure is typical of PVD thin films, but the unusual angular orientation of the grains seems to be a consequence of the deposition

Fig. 3. A cross section SEM image of Cu-doped Be sputter-deposited on a plasma polymer mandrel. This deposition required approximately 7 hours.

geometry. Note that the morphology is fairly well established within the first 5 μm or so. The outer surface of the film is quite rough, as shown by the SEM image in Fig. 4. AFM measurements of this surface give an rms roughness of about 250 nm. As suggested by the grain structure, the film has significant porosity. The calculated density is 1.78 gm/cm³, which, assuming 2 atom % Cu doping, is about 85% of full density. This estimate does not take into account other contaminants, such as oxygen, which are certainly present at some level. Preliminary studies suggest that this morphology is highly permeable to hydrogen, to the extent that the permeation is probably controlled by the polymer mandrel.

Fig. 4. An SEM image of the outer surface of the film from Fig. 3.

In order to verify that this deposition process could be used to produce full-thickness capsules, a 10-day run was performed, with the sources running for 8 hours per day. Initially, there were 10 capsules in the bounce pan. After two days, the chamber was opened and two of the capsules were removed. This procedure was repeated until the end of the run, yielding samples with 5 different film thicknesses. Capsules removed after 2, 4, and 6 days have measured thicknesses of 26 μm , 51 μm , and 80 μm . There is little qualitative change in the morphology of the cross sections as the thickness increases, although careful examination reveals a modest decrease in the size

of the grains. This is illustrated in Fig. 5, which shows a cross section of a capsule with an 80- μm -thick coating. Magnified views from the inside, middle, and outside of the film are shown to illustrate the consistency in microstructure. Over the course of the run, however, there was a change in the surface roughness. The capsules referred to above have rms roughnesses of 140 nm, 110 nm, and 73 nm, respectively. However, the two thicker films from this run were somewhat rougher: 110 nm and 91 nm. The initial reduction in surface roughness with increasing thickness (recall that the 13.4 μm film from Fig. 3 had an rms roughness of ~250 nm) may be caused by abrasion of the capsules against each other and the bounce pan.

Fig. 5. A cross section obtained from a capsule removed midway through an extended run. The deposition time was approximately 48 hours.

The roughness values obtained for the films described above are higher than acceptable for a NIF target capsule. Thus, a significant goal of this program has been to determine fabrication methods that result in smoother films. One approach we have started to investigate is that of applying a voltage bias to the bounce pan. This causes ambient ions to bombard the depositing film, which influences the film in a variety of ways. These include enhancement of surface atom

Fig. 6. a) A cross section from a capsule deposited with a 120 V bias applied to the bounce pan. The view is of the outside edge; the light colored area is the epoxy used to mount the capsule. b) An image from the same section as Fig. 3 but with the same magnification as (a).

mobility, sputtering of both Be and adsorbed contaminants, and implantation of argon, all of which can affect the morphology of the film. The initial series of experiments had bias voltages of 40, 80, and 120 V. The 40 V run showed little difference from the unbiased deposition. At 80 V, some disruption of the Be grain growth became evident, and the cross sections showed that the columns were oriented more perpendicular to the surface. At 120 V, the morphology of the film was completely different from the no-bias runs. Figure 6a shows a cross section from one of these films. For comparison, Fig. 6b shows part of the cross section from Fig. 3, but magnified to be at the same scale. Note the smaller and more densely packed grains in the bias film.

Figure 7 shows an image from the outer surface. The ion bombardment has inhibited grain growth to the extent that the grains are only a few tenths of a μm in diameter, as compared to the 1 to 2 μm grains seen in Fig. 4. These films appear shiny to the eye, and AFM measurements give an rms roughness of about 50 nm.

Although the films were deposited for 7 hours, they are only about 7 μm thick. This may be due to resputtering of the depositing film by incident ions, as well as the formation of a more compact film. The measured density is quite high ($\sim 2.8 \text{ g/cm}^3$), which suggests the presence of significant amounts of implanted Ar. This has been qualitatively verified using XRF. Experiments are ongoing to test whether post annealing removes this contaminant from the Be layer.

One of the significant advantages of the sputter deposition technique is the ability to produce arbitrary (within reason) dopant profiles. The results from a feasibility experiment are shown in Fig. 8, which shows the copper concentration as a function of distance from

Fig. 7. An image of the outer surface of the capsule from Fig. 6.

Fig. 8. A plot showing the measured concentration of Cu as a function of distance through a 60- μm -thick Be film. The zero of the abscissa refers to the inner surface of the film, i.e., adjacent to the mandrel. The data were obtained using electron microprobe analysis on a polished cross section of the film, shown in the inset photo.

the inner surface of a 60- μm -thick Be capsule. In this case, rather than the composite Be/Cu target mentioned above, one of the Be sputter targets had holes drilled in its face into which were inserted segments of copper wire. The power applied to the target was not changed during the first 40 μm of deposition, but since Cu has a higher sputtering rate than Be, the wires erode faster than the rest of the target. As the Cu wires recede below the surface, the Cu rate decreases, and eventually equals the Be rate. This produces the concentration gradient shown. After 40 μm of deposition, the gun with the Cu/Be target was turned off. In general, it would be a simple matter to vary the power applied to a Cu or Cu/Be composite target to produce an arbitrary profile.

DISCUSSION

The unbiased deposition experiments have shown that Cu-doped Be films of any desired thickness can be grown on polymer mandrels. The biasing experiments have demonstrated that the film morphology can be significantly modified by ion bombardment during deposition. The surface finishes of these films are rougher than will ultimately be required, so a priority of

future work will be to improve this property. Ion-assisted deposition (via either pan biasing or an external source) in particular is a promising avenue to continue exploring. A new deposition chamber is being constructed that will permit the use of an external ion gun concurrent with deposition. This will allow us to adjust the ion current, energy, and possibly species independent of the sputter sources. Another approach we have begun to study is the use of intermittent reactive gas dosing during deposition. Each pulse, which would typically be of only a few hundred milliseconds duration, forms a thin compound layer (e.g., BeO or Be₃N₂). Repeating this process every 50-100 nm promotes a fine grain structure by inhibiting the growth of Be grains. In the future we also intend to investigate the use of low-Z solid-state dopants such as boron to accomplish the same goal. Finally, it is possible that post processing of some sort, such as mechanical polishing, might be used to improve the surface finish of the as-deposited films.

As mentioned in the introduction, strength and permeability are also very important film properties. Strength measurements are just beginning on capsules from the extended run, and will become routine as more samples become available. Permeation studies are waiting for the construction of a suitable test station.

CONCLUSION

The experiments done to date have demonstrated that doped Be capsules similar to NIF designs can be fabricated using sputter deposition. Although many questions remain to be resolved as to strength, surface finish, and permeability of these films, promising results have been obtained with only preliminary studies of process modification. This provides strong motivation for continuing our efforts in this area.

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McEACHERN/697

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McEachern/7267