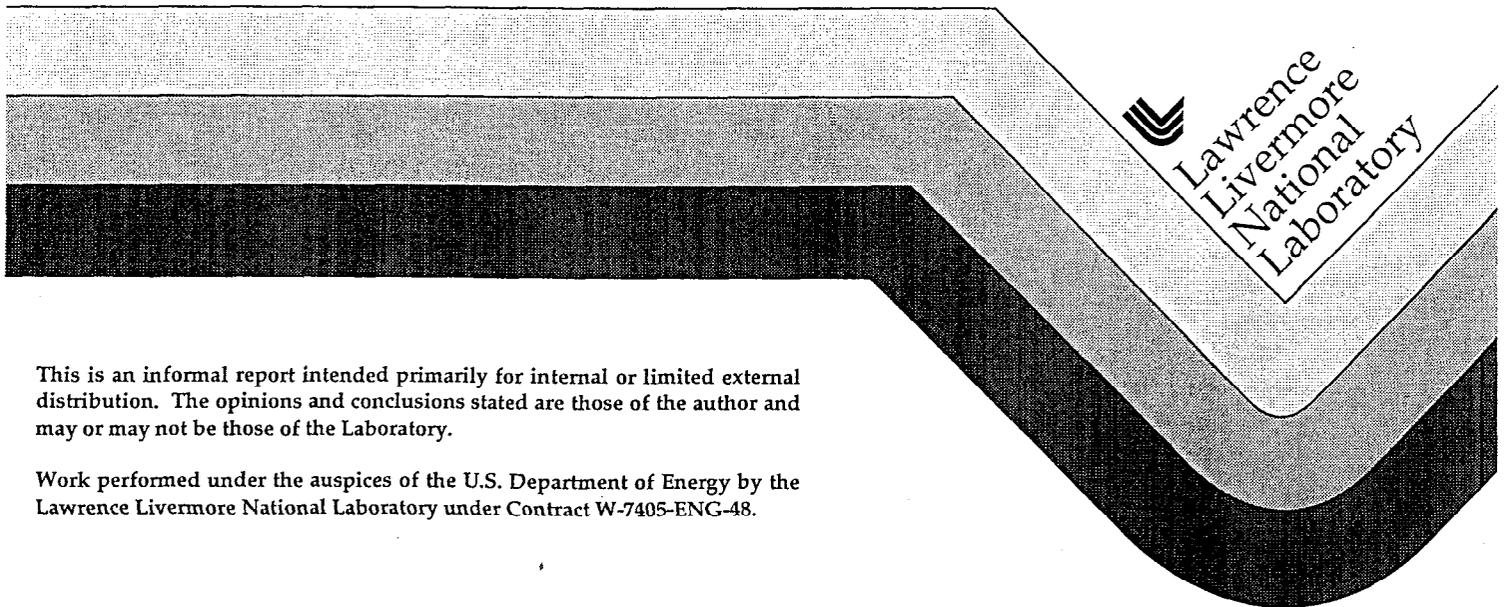


Tailoring Material Properties of Sputtered Beryllium

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Doped beryllium is a material of considerable interest to both the ICF and the weapons communities, as well as finding application in specialized industrial settings (e.g., x-ray windows and mirrors). Some of these uses require conformal coating of thin films on (possibly) irregularly-shaped surfaces. Physical vapor deposition (PVD) is often used to accomplish this, and sputtering is often the technique of choice. Among its advantages are that the depositing atoms are relatively energetic, leading to more compact films. Moreover, by simply applying a voltage bias to the substrate, ambient noble gas ions will bombard the growing film, which can cause further densification and other modifications to the microstructure. Sputtering is also well suited to the introduction of dopants, even those that are insoluble.

Most applications of these novel materials will require fundamental knowledge of their properties. Because so many can be devised, such information is generally unavailable. The objective of our effort has been to systematically study the properties of films produced under different conditions, with an emphasis on surface finish and permeability. We have made extensive use of atomic force microscopy (AFM) and electron microscopy to determine the microstructure of the films, along with composition probes (mainly x-ray fluorescence) to quantify the chemical structure.

Our studies can be roughly divided into three categories. First, there are those in which the properties of pure or Cu-doped Be films have been investigated, especially on randomly-agitated spherical capsules. Included are studies of the effects of a constant substrate bias ranging from 0 to 120 V and application of an intermittent bias during deposition. Second, there are experiments in which the structure of the depositing films has been modified via the incorporation of dopants, primarily boron. Finally, there have been numerous attempts to characterize the permeability of Be coatings at temperatures ranging from 200°C to 500°C.

Deposition of Be onto moving spherical substrates differs significantly from films on planar substrates. As with most metals, PVD-deposited Be tends to form a pronounced columnar grain structure, with the columns oriented normal to the surface. On planar surfaces, the grains tend to become larger with increasing film thickness, with a corresponding increase in the surface roughness. On capsules, however, the diameter of the Be grains is established within the first ~5 μm of coating (roughly 1 μm) and remains largely unchanged even for layers in excess of 100 μm thick. The surface roughness, as measured on a patch using AFM, also remains fairly constant or improves somewhat, with limiting values in the neighborhood of 100 nm rms. In an attempt to reduce the grain size and improve the surface finish, we studied application of a voltage bias to the

substrate during deposition. This had a significant effect on the morphology of the Be films. For 8- to 10- μm -thick films deposited on plastic spheres, the rms roughness decreased from ~ 150 nm with no bias to ~ 40 nm with a 120 V bias. At the same time, the grain size was reduced, and the film density increased (reflecting the elimination of voids). X-ray fluorescence measurements detected the presence of implanted argon in 120 V bias films, but not at 80 V bias or below. This observation is relevant for applications sensitive to the x-ray transparency of these coatings. Finally, two experiments were conducted using intermittent biasing: 1 min at 100 V/10 min at 0 V and 1 min at 200 V/10 min at 0 V. In both cases, the biasing appeared to have no significant effect. These results are documented in the accompanying article, "Sputter-deposited Be Ablators for NIF Target Capsules."

Another means of refining the Be grain structure is by introducing insoluble dopants. We have extensively studied the use of boron for this purpose. As a means of modifying material properties, this dopant is of particular interest for x-ray related applications by virtue of its low Z. We performed a series of studies on the effects of adding boron to Cu-doped beryllium films deposited on silicon flats. By varying the boron concentration in small increments between 5 and 18 atom %, we determined that there is an abrupt transition at approximately 11 at.% B from films with a roughness of ~ 20 nm rms (less boron) to ones with a roughness of ≤ 2 nm. Away from this transition, the roughness and morphology change only gradually with boron content. We have also studied B-doped films deposited on capsules. Samples with 3, 5, 10, 15, 20, and 25 at.% B have been produced and characterized. Several distinctive morphologies have been observed. At 3 and 5 at.%, the films have uniform, straight columnar grains with well-defined crystal facets at the surface. At 10 at.%, poorly-defined columnar grains are accompanied by rounded surface features. At higher boron concentrations, no structure is visible in SEM micrographs of fracture cross-sections, and the surfaces are characterized by rounded nodules varying in size up to ~ 1 μm . A boron concentration of 15 at.%, somewhat above the minimum required for a desirable bulk morphology, was chosen for further study based on the observation that films with high levels of boron were more prone to degrade at elevated temperatures. The second attached paper, "Evaluation of Boron-doped Beryllium as an Ablator for NIF Target Capsules," discusses these experiments in more detail. Coatings of varying thickness on capsules revealed that unlike our experience with undoped Be, this material becomes significantly rougher as the coating becomes thicker, exceeding 400 nm rms for a 15- μm -thick film. Experiments on stress disks and plasma-polymer-coated flats have demonstrated that B-doped Be films deposit with high levels of compressive stress and adhere poorly to the plasma polymer, a combination which may contribute to the dissimilar results on capsules and flats. Finally, we performed a limited number of experiments in which pulses of nitrogen were admitted to the vacuum chamber during deposition. This was to test the hypothesis that the layers of nitride thus formed would disrupt the growth of Be grains, leading to a more fine-grained microstructure. In these experiments, a 0.8 second pulse of N_2 was released in the vacuum chamber every 5 min (corresponding to ~ 150 nm of Be coating). The resulting films were smoother, with roughnesses of 60 to 70 nm rms, but were also highly stressed and quite brittle. Some of samples fractured spontaneously. For most applications, these characteristics would be highly undesirable, so these experiments were not pursued.

The majority of our permeability experiments have been performed at a temperature of about 200°C. This was chosen based on results from Los Alamos¹ in which thin palladium coatings on both sides of a Be foil were found to significantly increase its hydrogen permeability. The literature values² for the permeability of bulk Be do not predict permeation rates as high as those observed at LANL, but the large potential benefit and positive preliminary data motivated several attempts to reproduce this result. We tested this concept by depositing thin (~100 nm) layers of Pd onto plastic capsules, overcoating them with Be, then finishing with another thin layer of Pd. These capsules were kept in a heated chamber under high-pressure deuterium for several days, then crushed inside a calibrated volume to measure the gas fill. Some initial experiments demonstrated significant permeation, but subsequent, careful experiments showed no permeation, strongly suggesting that sputter-deposited Be does not have greatly enhanced permeability compared to the bulk-processed Be that is normally used in permeation experiments. Low-temperature permeation of B-doped capsules was also unsuccessful. At the same time, efforts have been made at General Atomics to measure permeability of Be coatings on glass mandrels in the 400° to 500°C range. This effort has thus far been complicated by the persistent appearance of a substantial oxide layer during the fill process, which prevents permeation. We are currently studying how to prevent this phenomenon.

- 1) R. Dye, R. Henneke, L. Salzer and L. Foreman; poster presentation at the 1998 Target Fabrication Specialists Meeting, Jackson Hole, Wyoming; April 20, 1998.
- 2) G. R. Longhurst, R. A. Anderl, T. J. Dolan, and M. J. Mulock, *Fusion Technology* **28**, 1217 (1995).

