PREPRINT UCRL-82778

Lawrence Livermore Laboratory

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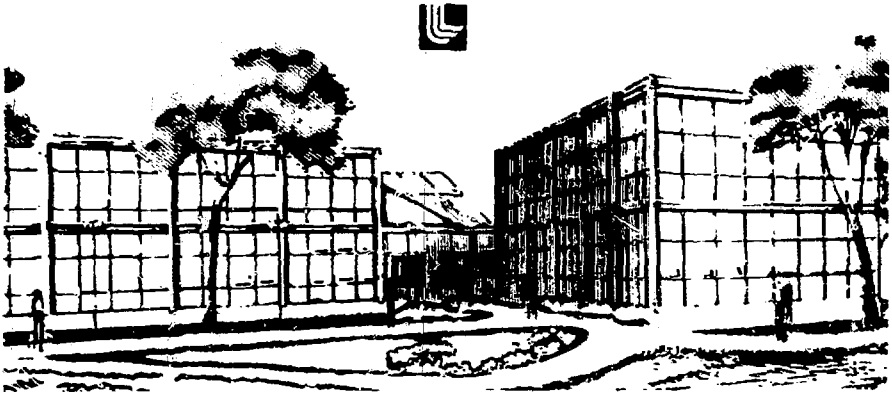
September 19, 1979

This paper was prepared for submission to
26th National Symposium of the American Vacuum Society
October 2-5, 1979, New York City



RF MAGNETRON SPUTTERING OF THICK FILM AMORPHOUS BERYLLIUM*

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ABSTRACT

Thick film coatings of beryllium, needed for the low-Z ablator layer in proposed laser fusion targets, have been prepared using high rate magnetron rf sputtering. The requirements for these Be coatings include thicknesses from 5 to 50 μm, complete freedom from surface defects, and an average surface roughness of 100 nm or less. These specifications are difficult to achieve if crystalline growth occurs during sputtering, since the surface roughness of thick grainy films can be on the order of micrometers. We have sputtered very smooth, dense, thick Be films with surface roughness less than 100 nm. X-ray diffraction analysis of impurity doped films indicates an amorphous-like structure. Impurity stabilized amorphous Be with smooth surfaces is reported on both cooled copper and higher temperature glass substrates. The sputtering parameters (substrate temperature, deposition rate, argon pressure, and impurity gas levels) affecting surface roughness and film structure are discussed in terms of SEM, AES, and x-ray diffraction results.

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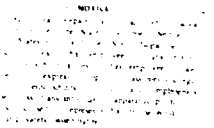
*Work performed under the auspices of the U. S. Department of Energy by the Lawrence Livermore Laboratory under Contract Number W-7405-ENG-48.



INTRODUCTION

Laser fusion targets designed for ablation driven implosions require low-Z, low density coatings as the ablator layer. The thickness uniformity and surface finish of these coatings must meet stringent specifications.[1] While numerous other studies of vapor deposited beryllium films have been made[2-4], none have yet reported the conditions necessary to deposit smooth, thick films on hot spherical glass substrates. We report here on development work at LLL on the fabrication of smooth, thick Be films that will be needed for the ablator layer on hollow glass microsphere (HGM) laser targets.

We first discuss the unique problems involved in coating HGM laser targets, and then describe our approach to achieving the required uniform, smooth, dense Be coatings on test substrates. Uniform coating on a sphere requires continuous motion during coating. The negligible heat conduction at the contact point of a moving sphere makes substrate cooling difficult. Despite this thermal isolation, the HGM temperature rise during coating must be ed to prevent damage to the high pressure DT filled HGM. These temperature limitations suggest a low incident energy, low power deposition; however, other constraints make low energy deposition undesirable. First, the high degree of oblique incident flux inherent in coating a spherical substrate promotes a porous, rough film growth, which can only be minimized by using a high incident energy, low pressure deposition process.[5] Second, the effective deposition rate from a single source onto a HGM surface is reduced by a factor of 4 to 6 compared to a planar substrate, and finally, the sputtering rate for Be is one of the slowest of any metal. As a

compromise we use a high energy, high power, low pressure rf magnetron sputtering process, and limit the power to that necessary to prevent temperature damage to the HGM. However, this still means coating onto a substrate (HGM) at $100^{o}C$ or more, where surface roughness from Be grain size is a problem.

In the following sections we present the sputtering parameters and film structure that result in fine-grained or amorphous thick Be coatings with smooth surfaces using isolated glass slides and cooled copper mandrels as simulated substrates.

EXPERIMENTAL

The rf sputtering system uses a cylindrical magnetron (Sloan S-310 Sputtergun) converted to rf operation using a detached matching network with a 1 kW, 13.56 MHz generator. The 20 cm dia by 20 cm high stainless steel diffusion pumped vacuum chamber has an ultimate base pressure of $1x10^{-5}$ Pa. A manual high vacuum valve is used as an adjustable throttle during sputtering. The normal throttled base pressure is $4x10^{-5}$ Pa. Sputtering pressure is normally servo controlled at 0.5 Pa with throttling adjusted to limit foreline pressure to 13 Pa, corresponding to a 45 SCCM flowrate.

Substrate fixturing consists of an electrically isolated, water cooled copper mounting block. All chamber surfaces exposed to the Be flux are protected by de-mountable, bead-blasted copper shields to prevent flaking during extended depositions, and to facilitate chemical cleaning afterwards.

The results presented here are obtained with the substrate mounting block dc grounded through an rf choke and ammeter. At the start of a run, the floating dc potential is noted, and then the grounded dc current monitored during the duration of the deposition. Typical substrate self-bias values at 1 kW rf, -300 V cathode bias, and 0.5 Pa argon are: +25 VDC floating and +100 mA grounded. With a target to substrate spacing of 4 cm, the Be deposition rate is 83 nm/min. A copper shutter is used to shield the substrates during a pre-sputter period. A 5 minute pre-sputter at 1 kW is sufficient to remove the oxide from the Be target. Extremely pure Be films are obtained with a 1 hour pre-sputter period to heat the Cu shielding and bake out the vacuum system. The strong oxygen gettering action of Be is also an effective pump during the pre-sputter period.

Substrates consist of glass microscope slides and 3 mm copper rods (mandrels) with one end metallographically polished to a 50 nm finish. The Cu mandrels are securely clamped into the mounting block with only the polished end protruding above the block. The glass slides are loosely set on the mounting block near the mandrels. All substrates are ultrasonically solvent cleaned to remove particulates and gross organic contaminants. The Cu mandrels are collodion stripped to remove adherent particulates, and then hydrogen plasma stripped to remove any residual organics. The glass slides are oxygen plasma stripped after the ultrasonic cleaning. The substrates are loaded into the sputtering system and pumped down while still warm from the plasma stripping.

Film analysis consists of interferometric step determination of thickness, SEM analysis of surface finish and crossection structure, Cu-Kα x-ray diffractometer scanning, and impurity determination with

Auger electron spectroscopy on a PHI spectrometer. The worst defect amplitude (WDA) as a measure of surface finish is defined from a SEM view of the peak-to-valley height of the worst defect.

RESULTS AND DISCUSSION

The Auger bulk impurity analyses for all deposited Be films ensure a reliable interpretation of film impurity to surface finish and structure. We feel this is the preferred analysis, instead of the more common statements of system impurity levels, e.g. residual gas analysis during or before sputtering. An example of the purity level possible in our Be films is shown by the top trace of the Auger electron energy scan of Fig. 1. Note that no detectable impurities are present in this sample, a 22 μm thick Be film deposited for 3 hours on Cu at 15°C, after a 1 hour pre-sputter. This scan was taken in the bulk film, 300 nm below the surface, with in situ argon sputter etching during AES analysis providing the small argon peak. The middle trace in Fig. 1 shows a typical Auger analysis in the bulk of a doped Be film containing 9 at. % oxygen. The bottom trace shows a surface scan of the same sample showing the nearly complete Be oxidation at the surface.

Dopant Effects on Heated Substrates

High purity (18 μm) thick Be films deposited on glass during the same run as the pure sample analyzed above, show large crystal facets and an unacceptably rough surface finish demonstrated by the SEM micrographs of

Fig. 2. The 40 μm dia highly faceted area of Fig. 2(a) may have resulted from a small contaminated area on the glass surface such as a dried water spot. Such microscopic contaminants can provide preferred nucleation sites for crystal growth, and result in the amplified surface features shown here. These large crystallites are an example of Zone 2 growth from the Thornton sputtering model.[6] The SEM photomicrograph of Fig. 2(b) shows the hexagonal Be {002} pyramid grains in the contaminated area. These {002} facets rise up to 1.5 μm above the smoother areas, and constitute unacceptable defects for laser fusion target coatings. The x-ray diffraction (XRD) scan of this film (Be-1) in Fig. 3(a) shows a strong {002} texture orientation, with a much weaker {110} orientation. Additional details for this sample are listed in Table I.

A film (Be-2) with oxygen, nitrogen, and carbon impurities totaling 2 at. % (see Table I) results in a marked improvement in smoothness, as shown in Figure 4(a). The strong {110} texture orientation is shown in the (XRD) scan in Fig. 3(b). The differences in surface structure between Fig. 4(a) and 4(b) are believed to be caused by a substrate temperature variation between the two films which were deposited in the same run. Crystal {002} facets have now grown out of the {110} background on the presumably hotter slide giving a much larger defect on the surface in Fig. 4(b). Since substrate temperatures cannot be controlled in actual HGM coating, it is important to avoid the crystalline grain growth problems illustrated in these examples.

Additional doping of the Be films with a gaseous impurity can suppress the crystalline growth process in heated Be films. Sample Be-3, shown in Fig. 5(a), contains 22 at. % bulk impurity (see Table I) from

doping with N_2 during deposition. Note the absence of a crystalline type surface structure. This film has a WDA of 280 nm. The XRD scan for this sample, Fig. 3(c), shows no Be lines, indicating this film has been N_2 impurity stabilized in an amorphous state. A similar result is illustrated with the oxygen doping of sample Be-5 in Fig. 5(b), where 14 at. % oxygen has improved the WDA to 160 nm. This film is also amorphous, with an XRD scan identical to Fig. 3(c).

We have also briefly investigated H_2 doping of a Be film by sputtering in an H_2 partial pressure of 1.6×10^{-2} Pa. Sample Be-8 in Table I summarizes the result. The WDA was 280 nm, with small mixed crystal facets contributing to the surface roughness. The XRD scan, Fig. 3(d), shows a nearly random orientation with the strong {002} texture of clean Be films nearly suppressed.

Low Substrate Temperature Effects

Results on water-cooled copper polished substrates held at 15°C (T/T_m=0.19) provide useful comparative information on the temperature dependence of grain size, surface finish, crystal structure, and impurity stabilization of amorphous Be.

From the Thornton zone model of sputtering,[3] both Zone 1 and Zone T growth could occur depending on pressure. Sample Be-9 in Fig. 6(a) (deposited at 300 W and 3.3 Pa) illustrates the rough, open, low-density Zone 1 growth, with a WDA of 560 nm. This film is also an unusual example of {101} texture orientation, shown in Fig. 3(f). The impurity level of the Be-9 film is similar to the Zone T structure of sample Be-11

(deposited at 1 KW and 0.80 Pa), which has a WDA of only 48 nm with a {100} orientation. This comparison is an example of a Zone 1 growth at high pressure vs a Zone T (smoother surface) growth at low pressure.

A 22 μm thick Be film surface, deposited at 0.49 Pa argon on 15°C Cu (Be-10 in Table I) is shown in Fig. 6(b). AES analysis found no detectable impurities in this film while the XRD scan of Fig. 3(g) indicates a 100% {100} orientation. This film has a fine-grained surface structure, with an WDA of 140 nm. The "X" channel in the center right of Fig. 6(b) is a replication of the Cu substrate polishing scratches through 22 μm of coating. This is a demonstration of fine-grained, dense, uniform columnar crystals typical of a smooth zone T growth.

The chief effect of impurity doping on the 15°C substrates is to shift the orientation from a weak {100} to a weaker {110} to finally an amorphous structure, with a slight concurrent improvement in the already extremely smooth surface (Figs. 7 and 8). The low intensity of the only diffraction peak, (100), suggests that sample Be-11 with 1.5 at. % impurity has a highly disordered small grain structure with an improved surface finish as shown in Fig. 7(a). The very weak (110) Be line in Fig. 3(h) indicates that sample Be-12 (3.7 at. % impurity) is mostly amorphous with a further improvement in smoothness as observed in Fig. 7(b). Sample Be-13 with 9 at. % oxygen impurity is completely amorphous with a WDA surface of 36 nm as shown in Fig. 8(a). The fracture cross section SEM micrograph in Fig. 8(b) shows no apparent crystalline structure for this film.

SUMMARY AND CONCLUSIONS

1.) With low pressure, impurity-free rf sputtering and substrate temperatures above 200°C, we obtain large-grain, rough surface, high purity Be films with a strong {002} texture orientation and a weaker {110} matrix orientation.

2.) Impurity gas doping during sputtering changes the orientation and improves the surface finish. At 1-2 at. % impurity the orientation shifts to a {110} texture and surface finish improves to 360 nm WDA.

3.) At higher impurity levels, 5-20 at. % O_2 or N_2, the Be film is amorphous, apparently impurity stabilized. A very smooth finish of 160 nm WDA is possible at 200°C deposition temperatures.

4.) H_2 doping with hot substrates does not destroy the crystallinity, but changes the texture to a random orientation, with a surface finish of 280 nm.

5.) When Be is deposited at 15°C, the surface finish improves from 140 nm to 36 nm with increasing impurity content. Crystallographic orientation varies from strong {100} for high purity films, to weak {110} for 2-3 at. % impurity, to amorphous with over 5 at. % impurity.

6.) A low total sputtering pressure, < 1 Pa, is necessary to obtain dense, smooth Be coatings at low temperatures, in agreement with the Thornton zone model.

We conclude that low pressure, high energy rf sputtering with controlled impurity levels of O_2 or N_2 is a feasible technique to improve the surface finish of thick Be coatings needed on hollow glass microsphere laser fusion targets.

ACKNOWLEDGMENTS

The authors acknowledge the helpful discussions with C. M. Ward concerning the Auger analysis. We also thank V. K. Chen, K. L. Montgomery, and F. J. Wittmayer for their contributions in obtaining the SEM, Auger, and diffraction data, respectively.

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TABLE I. Properties and deposition conditions of rf magnetron sputtered thick Be films.

SAMPLE NUMBER	SUBSTRATE MATERIAL	FILM THICKNESS (μm)	P_I (Pa)	P_S (Pa)	TEXTURE ORIENTATION	FILM IMPURITIES (at. %) O	N	C	WORST DEFECT AMPLITUDE (nm)
Be-1	Glass[d]	18	4.0×10^{-5}	0.49	{002}	ND[e]	ND	ND	1060
Be-2	Glass	15	6.7×10^{-5}	0.57	{110}	0.5	0.5	0.9	360
Be-3	Glass	5	1.1×10^{-2}	0.67	a-Be[f]	1.7	20	0.7	280
Be-4	Glass	5	1.1×10^{-2}	0.67	a-Be	3	19	1.4	420
Be-5	Glass	5	5.3×10^{-3}	0.40	a-Be	14	ND	0.9	160
Be-6	Glass	5	6.7×10^{-3}	0.67	a-Be	5.5	11	1.5	220
Be-7	Glass	5	6.7×10^{-3}	0.67	a-Be	13	12	1.5	420
Be-8	Glass	9	1.6×10^{-2}[g]	0.61	Random	0.3	ND	1.0	280
Be-9	Cu[h]	5	1.1×10^{-2}	3.33	{101}	0.8	0.5	1.0	560
Be-10	Cu	22	4.0×10^{-5}	0.49	{100}	ND	ND	ND	140
Be-11	Cu	5	6.7×10^{-5}	0.80	{100}	0.5	0.3	0.7	48
Be-12	Cu	5	1.3×10^{-2}	1.33	a-Be + wk{110}	1.9	0.8	1.0	40
Be-13	Cu	5	5.3×10^{-3}	0.40	a-Be	9.2	ND	1.0	36

a - P_I = Total of Impurity Gas Partial Pressure.

b - P_S = Total Sputtering Gas Pressure = $P_{Ar} + P_I$.

c - AES determined bulk impurity content, at. % in Be.

d - Glass substrate temperature = 200°C ± 100°C.

e - ND = Non-Detected with AES analysis

f - a-Be = amorphous Be.

g - H_2 partial pressure.

h - Cu substrate temperature = 15°C ± 1°C.

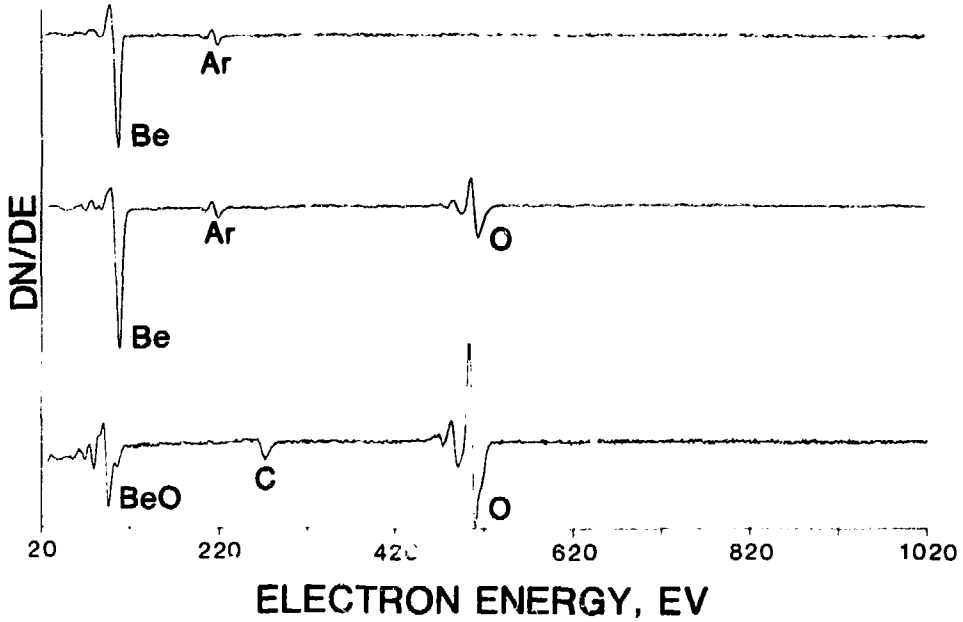

FIG. 1 Auger electron energy spectrums of a high purity, thick Be film (Be-10) sampled 300 nm into the bulk (top scan); a Be film containing 9 at. oxygen (Be-13) sampled in a bulk region (center); and the as-received surface scan of Be-13 showing the BeO surface layer (bottom).

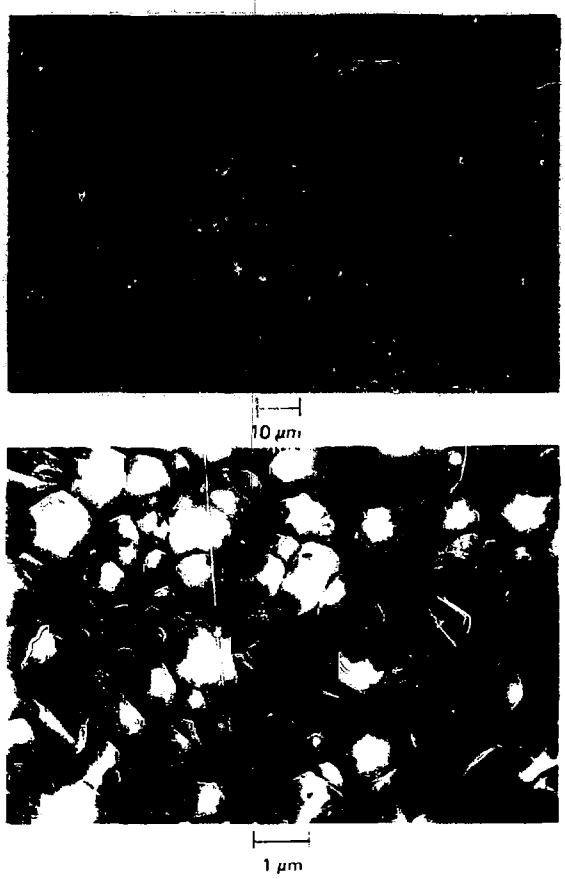

FIG. 2 SEM micrographs of a high purity Be film (Be-1) deposited on a hot glass substrate, (a) showing the faceted {002} crystallites growing from a contaminated substrate area, and (b) a top view at higher magnification of the rough crystalline area in (a).

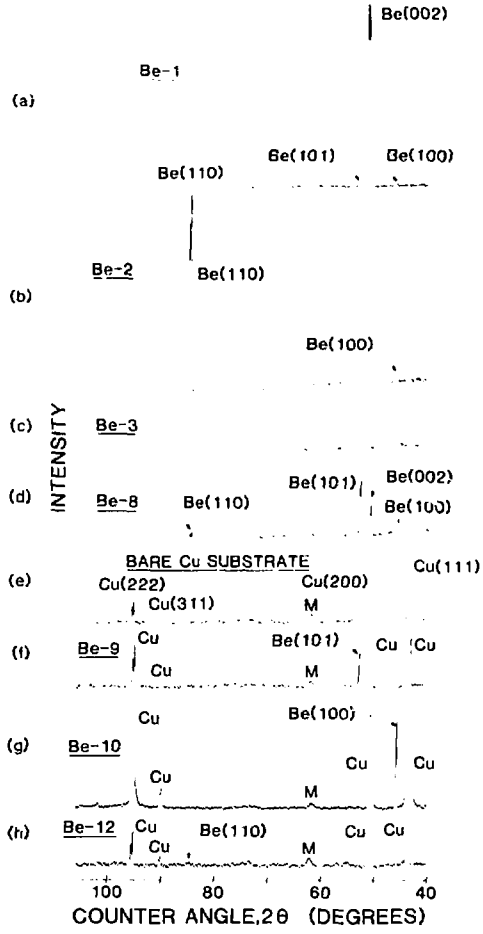

FIG. 3 X-ray diffractometer scans of Be films. See Table I for deposition conditions and impurity contents. Note: peak "M", occurring in Cu substrate scans only, is from the mounting fixture, e.g. see scan (e).

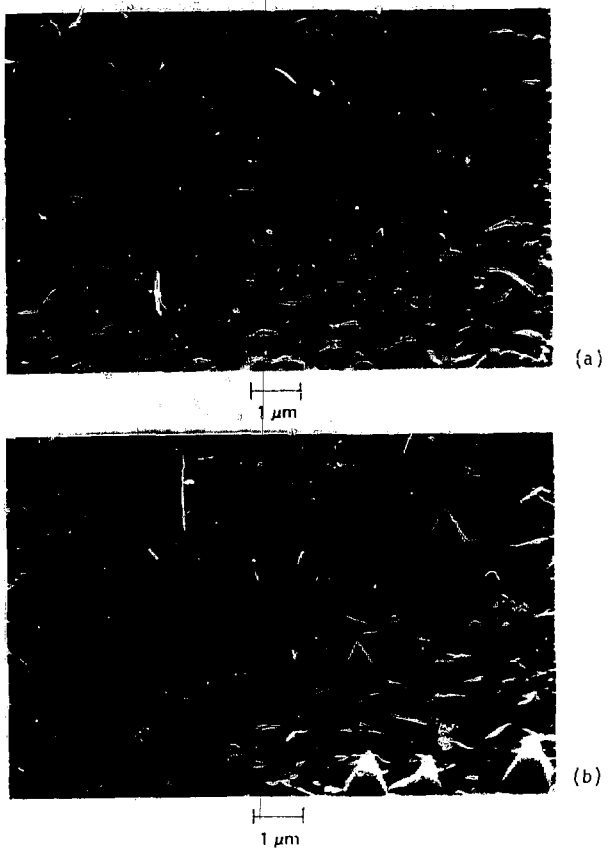

FIG. 4 SEM micrographs of the surface of Be films deposited on hot glass substrates (a) Be-2, with {100} orientation and 2 at. total impurity, and (b) a film from the same deposition as (a) where a higher substrate temperature has led to {002} crystallites on the surface.

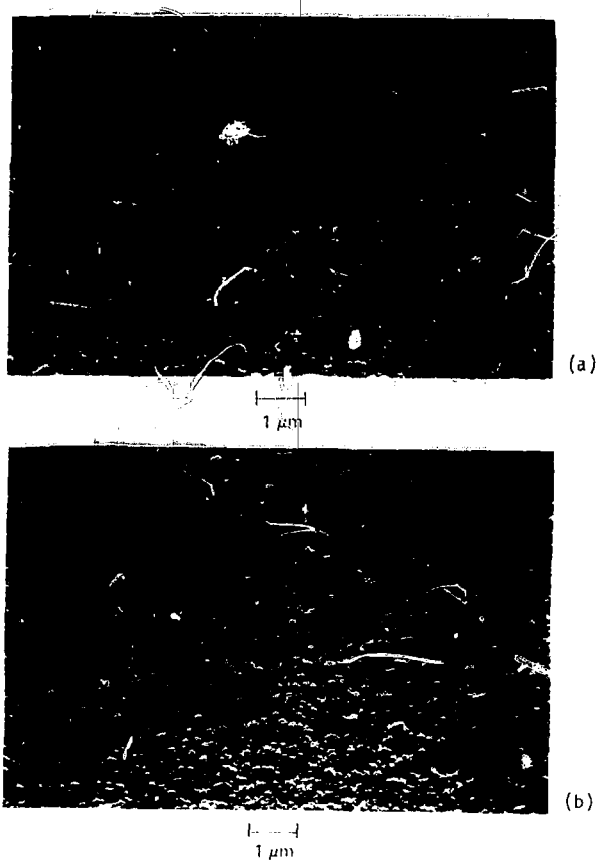

FIG. 5 SEM micrographs of impurity stabilized, amorphous Be films deposited on hot glass substrates: (a) Be-3, containing 20 at. % nitrogen; and (b) Be-5, containing 14 at. % oxygen.

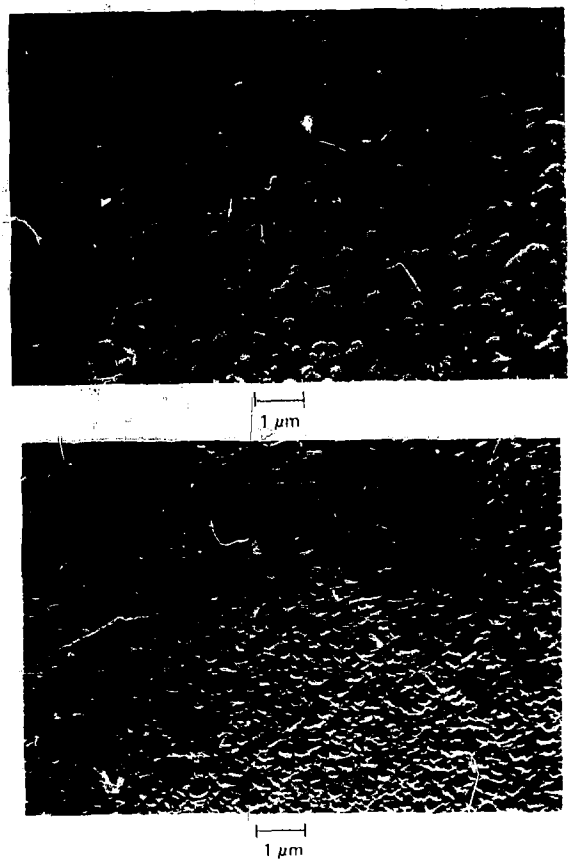

FIG. 6 SEM micrographs of Be films deposited at 15°C on Cu: (a) Be-9, showing rough growth from high pressure sputtering (Zone 1), and (b) the surface of Be-10, a high purity, 22 μm thick film (Zone T). Note the replicated Cu mandrel polishing scratches in the center right.

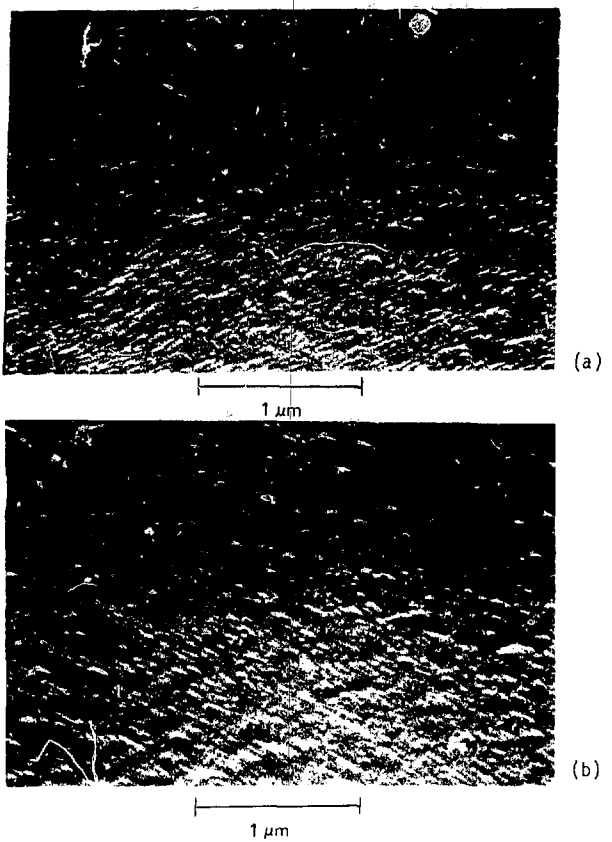

FIG 7 SEM micrographs of Be films deposited at 150C showing improved surface finishes with impurity doping: (a) Be-11, 1.5 at. % impurity, and (b) Be-12, 4 at. % impurity.

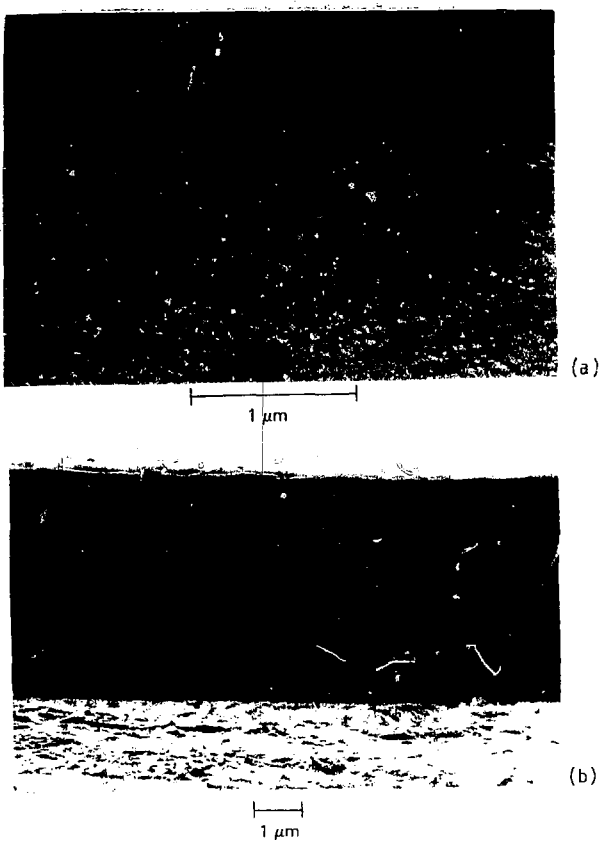

FIG. 8 SEM micrograph of Be-13, an amorphous Be film doped with 9 at. % oxygen, deposited at 15°C on Cu: (a) The smooth surface structure at 30,000X, and (b) a fracture cross section.