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2.B Aluminum/Aluminum-Nitride Sputter Deposition on the Inertial Fusion Target Using the Pulsed-Gas Process

Typical inertial confinement fusion targets consist of hollow glass micro-balloons (GMB's) of submillimeter diameter filled with pressurized DT gas. They are often coated with organic, and/or metal layers, and for successful implosion of these targets, high spherical symmetry and uniformity of the coating are essential. Variation in thickness and density of the coated layer should be less than a few percent and the rms surface finish less than 500 Å. Conventional techniques for metal coating using sputtering or evaporation do not provide these criteria due to the inherent, oblique incidence of the coating flux and poor control of substrate temperature in these processes. Depositions of aluminum have particularly poor surface morphology and low bulk density due to the columnar crystal growth[1] of an aluminum film.

A number of techniques have been extensively investigated as methods of obtaining smooth surface finishes and sufficient density uniformity of metal films. These include biased magnetron sputtering,[2] ion cluster beam coating,[3] and pulsed-gas sputtering processes.[4] Biased sputter coating has been employed in the coating of metal pusher layers onto inertial fusion targets to improve surface morphology. Ion cluster beam deposition also produces smooth, dense coating layers by accelerating an ionized cluster of loosely bound atoms into a substrate. This process has yet to be adopted for coating GMB's. Periodic pulsing of N_2 or O_2 during the sputtering process has been shown to form alternating layers of metal and metal nitride or metal oxide.[4] This technique suppresses columnar growth, produces high-yield-strength materials, and provides smooth surface finishes. To date this method has been applied to flat and cylindrical substrates, but we are unaware of any previous application of this technology to metal layer coatings on GMB's.

The present manuscript describes the application of the pulsed-gas process to coat pusher layers into glass microballoons to be used as fuel containers in inertial fusion experiments at LLE. In our experiment, aluminum was chosen as the coating material in the magnetron sput-

tering. An aluminum coating generally lacks high bulk density and displays rough surfaces, with frequent voids in the film. The effect of pulsed-gas processes in aluminum coatings is very pronounced.
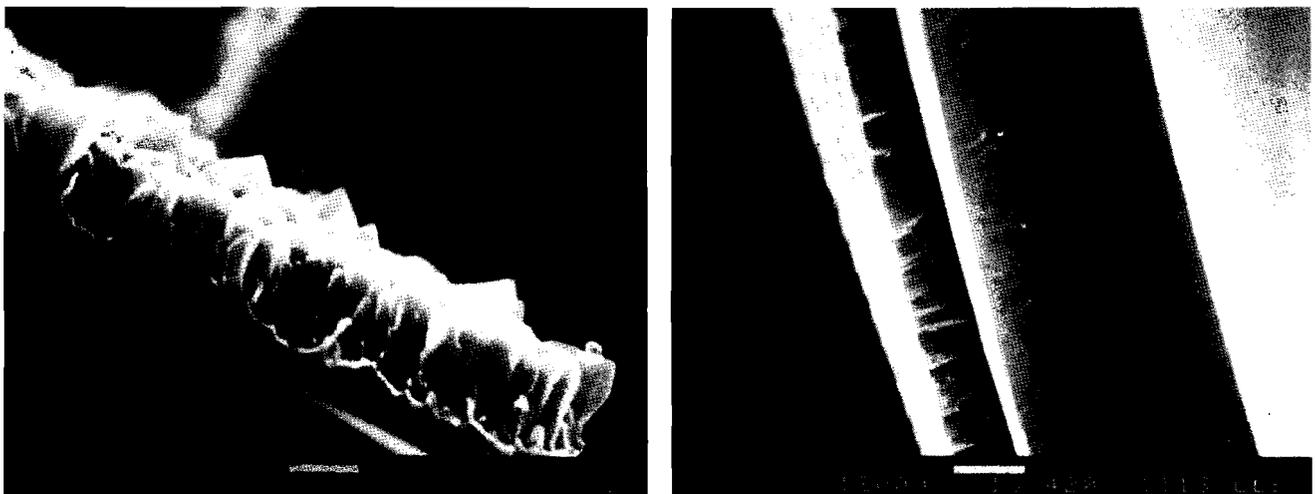
Deposition of aluminum coatings was performed in a vacuum system consisting of a stainless steel chamber pumped by an Edwards cryo-baffled 15-cm diffusion pump. The sputtering source was a Sloan S-310 magnetron sputter head. Argon sputter gas was purified in an R. D. Mathis gas purifier, and dry nitrogen was supplied from a liquid nitrogen gas phase line. An MKS-260 pressure controller was used with MKS-260 valves, an MKS-260 flow transducer, and an MKS-227 Baratron pressure gauge. Base pressure prior to coating was in the range of 10^{-4} Pa. The argon and nitrogen flows were set at 3.0 sccm, and the magnetron sputter head was operated at a dc current of 1.0 A. The nitrogen pulsing period was varied depending on the thickness to be coated. For coatings less than 2-μm thick, pulsing for 25 s every 3 min was satisfactory, that is, the density of the film and the surface finish were much improved compared with the aluminum coating without pulsing. During the nitrogen pulsing period, the flow rate of N_2 was kept as 3.0 sccm. For coatings up to 5 μm in thickness, pulsing for 2 s every 15 s was sufficient. At lower pulsing rates, the film tended to peel from the substrate.

Pulsed-gas processes have successfully been applied to produce high-yield-strength materials by forming alternating thin layers of high- and low-shear modulus materials. The effect of thin alternating layers is to inhibit the dislocation formation and the mobility of the dislocations, thus providing for improved mechanical properties. In addition, effective periodic inclusion of impurities as a result of this process improves specularity of the film surface substantially; it is this property that is of the utmost importance to us.

The effect of nitrogen pulsing on aluminum sputter coatings is shown in Figs. 21.13(a) and 21.13(b). Figure 21.13(b) is a scanning electron

Fig. 21.13
Scanning electron micrographs of sputter-deposited aluminum film (a) and aluminum deposit using a pulsed-nitrogen process (b). Frequency of pulsing was 25-s injections at 3-min intervals. Film density and surface finish are shown to be much improved by the pulsing.
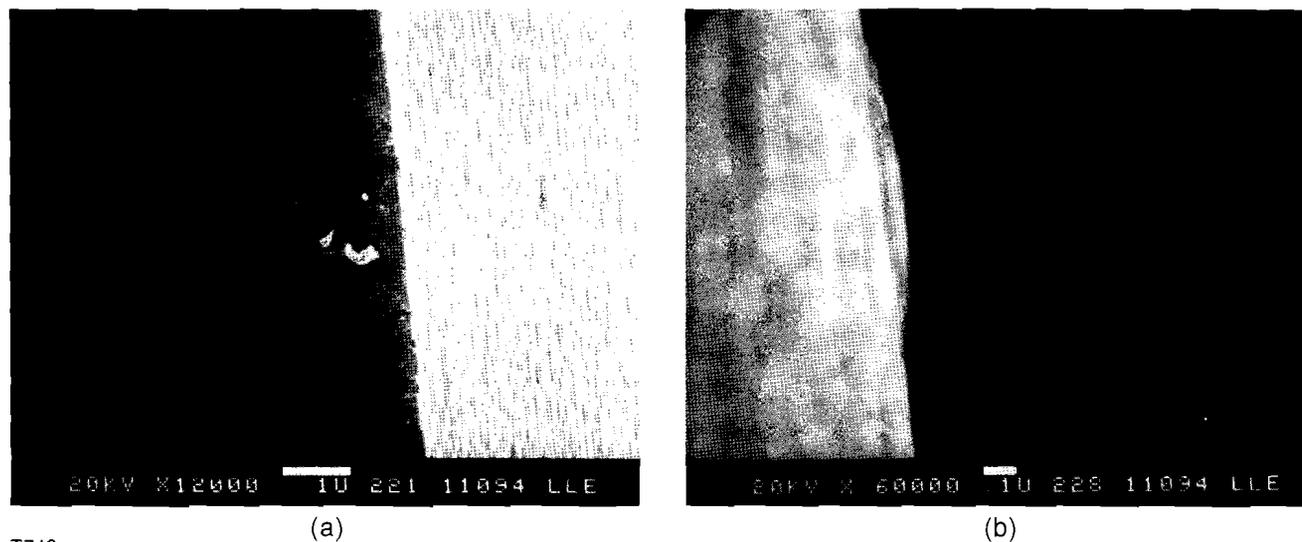

T667   (a)   (b)

micrograph of a 1-μm-thick Al coating that was obtained with N_2 pulses of 25 s injected at 3-min intervals. Compared with Fig. 21.13(a), which shows the structure of pure aluminum deposit, the effect on surface finish and on the removal of voids is dramatic. A careful examination of these photos reveals that the structure obtained with the N_2 pulsing process is not columnar, in contrast to pure deposit, nor lamellar. This observation is at variance with the published reports that describe a formation of alternating layers of pure deposit and aluminum nitride. Under conditions in the present experiment, the coating rate is 140 Å/min. A 25-s N_2 pulse duration at 3-min intervals implies deposition of alternating layers of 58-Å-thick aluminum nitride and 360-Å-thick aluminum. These dimensions are too small to be measured by a scanning electron microscope. Moreover, the profile of nitrogen injection is not a step function but one resembling a sinusoidal shape. The net result is to make deposition of nitrogen atoms more or less uniform into the aluminum matrix.

The surface finish does not appear to depend on the N_2 pulsing period. Figure 21.14(a) is surface morphology when N_2 gas was pulsed for 2 s in 12-s intervals. The surface finish of Fig. 21.14(a) is qualitatively the same as that of Fig. 21.13(b), which was obtained at a pulsing rate of 25 s in 3-min intervals. The amount of nitrogen gas injection is the same in both cases but the injection rate was different. A grain in Fig. 21.14(a) is magnified in Fig. 21.14(b), where its fractured cross section is magnified to show that the height of the grain is less than 0.1 μm.

Fig. 21.14
Aluminum film at nitrogen pulsing of 2 s in 12-sec intervals. Left photograph (a) shows an edge view of the film, while the right photograph (b) shows peak-to-valley dimensions of a major defect to be ≈ 50 nm.

(a)

T740

(b)

The structure and composition of the N_2-pulsed aluminum coatings were studied by x-ray diffraction and an energy-dispersive x-ray analysis technique. The x-ray diffraction scan was obtained by a Riguku diffractiometer; the results are shown in Fig. 21.15. Also depicted in Fig. 21.15 is the stick diagram of aluminum nitride, AℓN. The diffraction peaks are very shallow except for the one that appears at $2\theta = 36.7°$. Furthermore, comparison with the stick diagram of AℓN shows that the broad peaks of the diffraction pattern coincide with the combination of the peaks in the diagram. This implies that the main component of the
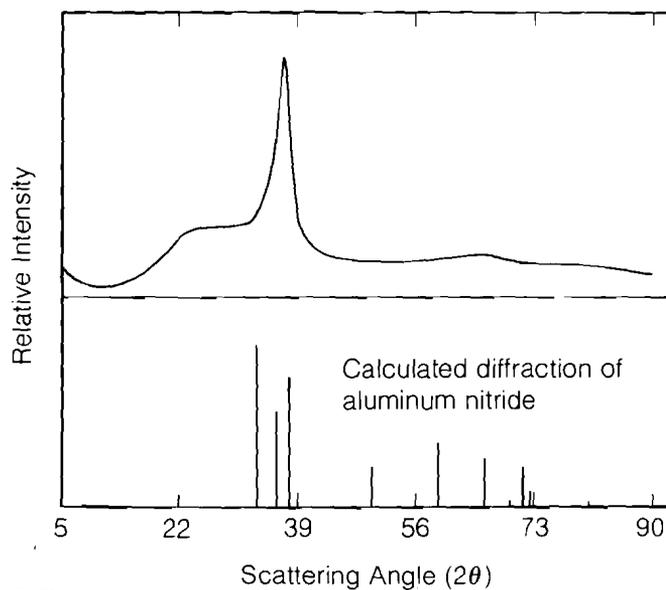
Fig. 21.15
X-ray diffraction pattern of a pulsed Al coating. The lower figure shows a stick diagram of aluminum nitride standard.

N_2-pulsed aluminum coating consists of very small crystallites of aluminum nitride. The broad peak near $2\theta = 22°$ may be that associated with the amorphous phase.

The chemical composition of the film was studied by an energy-dispersive x-ray analysis method using an EDAX unit. Figure 21.16 shows the energy-dispersive analyses of aluminum coatings with and without nitrogen pulsing. These two peaks are normalized to Al peaks. As expected, the presence of N_2 was detected from the scan of the nitrogen-pulsed peak. The content of nitrogen incorporation is believed to be less than 5 atomic %. The small oxygen peak appearing in both scans is due to the oxidation of the film when exposed to air.

In the pure Al deposit without N_2 pulsing, the oblique incidence of the flux of sputtered aluminum atoms causes self-shadowing, surface roughness, and voids, thus forming brittle coatings. Further, columnar growth, a characteristic crystal growth habit of thin-film deposition, also contributes substantially to the poor surface finish and inferior mechanical properties. When nitrogen ions, which can be considered defect-forming impurities, are periodically injected onto growing crystallographic planes, they poison the growth; therefore, ordinary dendritic crystallization cannot proceed. Deposit of nitrogen ions may become new heterogeneous nucleation sites. Under such conditions, either grain refinement occurs or an amorphous structure forms.

Although the injection of nitrogen gas is by perodic pulsing, it is not an abrupt on-and-off switching but resembles a sinusoidal shape. The net result is that nitrogen incorporation is more or less continuous. This precludes the possibility of forming alternating layers of aluminum and
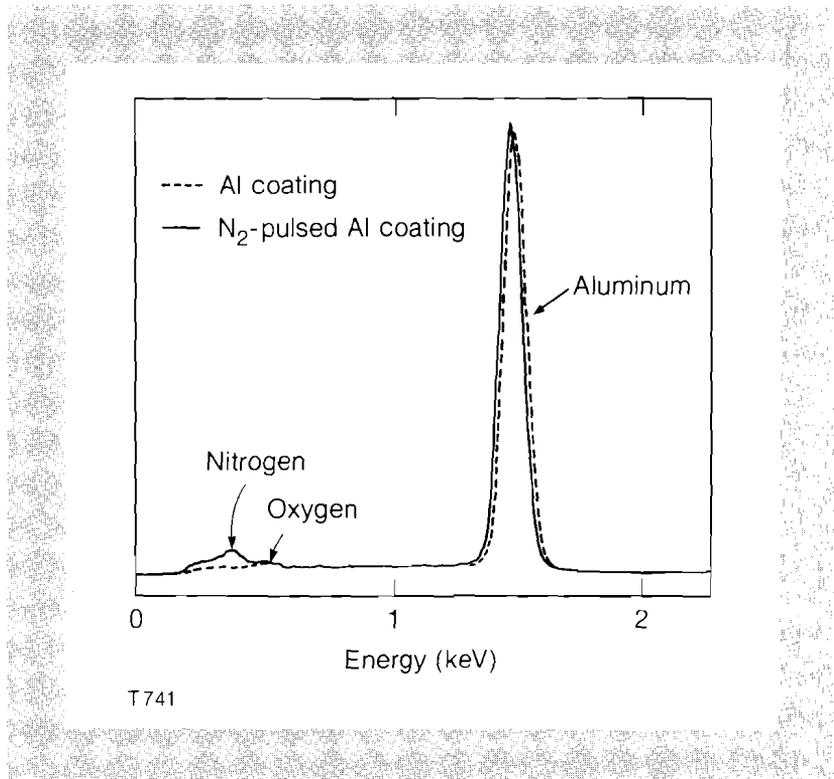
Fig. 21.16
An energy dispersive spectroscopy analysis of both pulsed aluminum and unpulsed pure aluminum. The pulsed sample is shown as a solid line; the unpulsed, a dotted line. The pulsed sample was normalized to pure aluminum peak.

aluminum nitride. It has been reported that epitaxial single-crystal growth of aluminum and aluminum nitride layers is a structure obtained by aluminum pulsing process and that this structure is responsible for high-yield strength. However, Fig. 21.16 indicates that the amount of nitrogen ions incorporated into the film is small, and our x-ray studies show that the structure is essentially amorphous. It is therefore concluded that the improved surface finish we obtained is due to the amorphous structure of the film.

ACKNOWLEDGMENT

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