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A SIMPLE SPUTTERING SYSTEM FOR
NUCLEAR-SCIENCE APPLICATIONS*

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September 1976

ABSTRACT

A sputtering apparatus with in-situ monitoring of deposit thickness is described. Its use in preparing thin foils and in reducing the thickness of commercially available foils is discussed in conjunction with a treatment of the system parameters.

* This work was done with support from the U.S. Energy Research and Development Administration.

INTRODUCTION

Fabrication of uniform films from high-melting-point or refractory substances is often difficult to realize with standard vacuum evaporators. Glow-discharge sputtering — that is, ejection of material from a surface by transfer of momentum from energetic ions formed in a glow discharge — has general applications in material removal and deposition, with the advantages of surface coverage and adhesion better than those afforded by vacuum evaporation.

Built from components readily available in the laboratory environment, the compact, low-cost sputtering system described here has found many uses in nuclear-reaction studies. The device has been used to make targets through sputter and reactive-sputter deposition and to reduce the thickness of stock foils for use as backing or degrader foils.

EXPERIMENTAL APPARATUS

A direct outgrowth of the apparatus described by Bolon¹, the sputtering-electrode assembly (fig. 1) is housed in a bell jar (the inverted base of a vacuum desiccator) evacuated by a 10 liters/sec turbomolecular pump with a liquid-nitrogen cold trap. As shown in the block circuit diagram (fig. 2), a 3 kV/40 mA voltage-regulated power supply (with a 40 k Ω series resistance to limit current) is used in this system in conjunction with meters to monitor voltage and arc current and a strip-chart recorder, which provides the history of the discharge current for sputter rate determination. Hastings gauge model SV-1C with a DV-3M tube is used to monitor pressures from one atmosphere to approximately 1 mTorr. Hastings Model SL-1 with type

-3-

DV-8 tube registers in the 10 to 0.01 mTorr range. Unless otherwise noted, all pressure readings given in this paper are taken directly from the gauges and not converted for the sputter gas. Table 1 indicates the pressures (Hastings gauge readings calibrated for air and corrected for an argon atmosphere), voltages, current, and dark zone values for several experiments.

Table 1

Pressure air	(mTorr) argon	Applied Voltage (power supply)	Discharge Voltage	Current (mA)	Dark Zone (± 0.5 cm)
17	30	3000	2600	8	7.5
50	80	3000	2100	21	4.0
150	260	1500	950	13	2.0
200	400	500	400	3	1.5
300	650	500	500	4	1.0

Interchangeable parts in the cathode (figs. 1 and 3a) allow for considerable latitude in application. Selective conductive shielding within the discharge dark zone will facilitate differential sputtering of the cathodic material and may provide a degree of collimation to enhance the line-of-flight (versus diffusion) component of deposition. With the discharge thus restricted, a defined portion of the cathode foil is preferentially removed, and contamination due to sputtering of mounting brackets, support framework, etc., is minimized. The cathode assembly may be moved vertically along aluminum posts. Changing the

orientation of the electrodes affects the geometry and mechanism of material deposition.

Sputtering progress is monitored in situ by measuring with a surface-barrier silicon detector (see fig. 3b) the energy loss of alpha particles of known incident energy.* Information on cathode or deposit thickness may thereby be conveniently obtained without having to remove the material for weighing (with the concomitant risk of breaking the foils). Also, the full width at half maximum of the degraded alpha-particle distribution serves as an indication of the quality of the film. The broader peak corresponds to particles sampling greater variations in foil thickness. Alpha-particle sources are located behind the cathode and on a mechanical arm which may be remotely moved to position the activity over the sputtered film. The glow discharge is extinguished prior to making thickness determinations. In order to protect the solid-state detector from being damaged by the energetic secondary-electron flux generated in the discharge, it is enclosed in a well-grounded conductive cage sufficiently thick to stop electrons in the keV energy range. Additional protection can be provided by a biased grid in the vicinity of the detector unit.

The argon sputtering gas is metered into the bell jar through double needle valves at a rate sufficient to maintain an ambient

*The 8.78 and 6.05 MeV alpha particles from ^{212}Po and ^{212}Bi , respectively, are used in this work. These activities are daughters of ^{212}Pb collected on (1" diameter x 0.005" thick) aluminum disks following the evolution of ^{220}Rn from a ^{228}Th source.

pressure of from 10 to 300 mTorr. Though industrial-grade argon is of sufficient purity in the majority of cases, at the higher pressures, contaminants in the gas and residuals in the system (in particular, water vapor and hydrocarbons) show a propensity to react with — generally oxidize — the more reactive sputtered materials.*

MATERIAL REMOVAL

Research applications often require metal foils of well-defined thickness. Energy degradation of heavy-ion beams, for example, is frequently achieved through the use of one or more foils of specific thickness. Very thin foils are preferable as target-backing material to minimize the heating of the target and the energy loss of the beam. The sputtering process provides a convenient way to reduce the thickness of commercially available foils for such purposes. The sputter apparatus described here has produced thin foils of good uniformity limited primarily by the crystal structure and morphology of the initial target material.

Sputter-gas ions, generated principally in electron-atom collisions²⁾ $e^- + G^0 \rightarrow G^+ + 2e^-$, accelerated across the cathode dark space impact on the cathode and may — if they are of energy in excess of the sputtering threshold of 20 to 30 eV — sputter target atoms through momentum transfer via elastic collisions. The lattice orientation of the cathode material is to be considered, because channeling

*Some difficulty was encountered in preparing films of molybdenum metal, due to its reactive nature. Agents desorbed from the sputter-chamber surfaces, backstreaming from the vacuum pump, and present as impurities in the sputter gas would often yield molybdenum oxides on reacting with the sputtered atoms.

of the incident particle may remove it from the surface where it is most effective as a sputtering agent. "Sputter focussing" (preferential material removal responsible for microscopic surface discontinuities) may result from correlated collision sequences propagated along crystallographic axes^{3,4}). Also, atoms situated at step edges or lattice defects are generally more easily removed. Nonuniform sputtering of material may therefore take place with cathode substances of regular lattice structure.

The more random atomic distribution in amorphous or polycrystalline solids, however, shows more uniform material removal in a glow discharge⁵). Sputtered atoms returned to the cathode surface following hard-sphere collisions, Penning ionization ($M^0 + G^* \rightarrow M^+ + G^0 + e^-$), or electrostatic interactions⁶) will not have the same degree of crystal order on redeposition and, hence, will not be subject to differential sputtering of the same magnitude. Increasing the operating pressure will enhance both the extent and spatial distribution of atom redeposition as the particle mean free path is decreased. (A decrease in redeposition is largely responsible for an observed doubling of the net removal of gold as the pressure drops from 40 to 20 mTorr.)

While foil thickness may be continuously reduced from the stock value with this technique, a practical lower limit for thickness is about 1 mg/cm^2 . Targets of well-defined crystal structure may show a high density of pinholes below this value, although sputtering at lower voltages (in the range of 0.5 to 1.0 kV) and higher pressure may inhibit pinhole formation.

MATERIAL DEPOSITION

The traditional application for glow-discharge sputtering is material deposition. In this case, atoms removed from the cathode migrate across the electrode gap to condense on the (anode) substrate. The properties of the resulting film are, to an important degree, determined by the system parameters during the sputtering process.

The ambient discharge-gas pressure affects the mechanism of atom deposition. At lower system pressures, line-of-flight transfer occurs, whereas a diffusion mechanism prevails for pressures at which the particle free-path length is much smaller than the electrode separation. A large diffusion component will, in general, yield relatively dispersed films with somewhat inferior adhesion to the substrate.

Elevated temperatures and substrate bias may be induced and sustained by an energetic secondary-electron flux. Electrically isolated substrate stages may attain potentials up to -1 kV during dc sputtering⁷). These potentials may markedly change film properties. (For example, observed film densities may be less than 50% of the theoretical.)

There is a rough proportionality between sputtering yield (the number of atoms ejected from the bombarded surface per incident ion) and ion energy. The sputtering rate, therefore, monotonically increases with the cathode-fall potential difference. The yield for argon, however, does not appreciably increase — and may decrease — for ions in excess of 5 kV energy⁸), because the ions implant too deeply to sputter effectively. The optimum discharge voltage, as with

pressure, is clearly determined by the task to be executed (see sections on Material Removal and Included Sputtering Gas). Attention to the cathode current density, a combination of ion and secondary-electron currents, will facilitate reproducible sputtering.

Data on sputtering rates for different metals at fixed applied voltage (3 kV) and pressure (40 ± 5 mTorr) are presented in table 2. There is good proportionality here with other tabulations⁹).

Table 2
Sputtering rates.

(V=3000 volts, P= 40 ± 5 mTorr)

Metal	mg/A-h ($\pm 20\%$)
Be	8 ^a
Al	40
Ni	350
Mo	90
Pt	2325
Au	5050

^aIncluded here for purposes of comparison, this value has been scaled from an electrode configuration somewhat different from that presented here.

At lower rates of material deposition and local temperatures sufficiently high for thermal migration of the atoms, the condensing film may show epitaxial growth, adopting the structure of the substrate matrix. An example of this phenomenon is the deposition of

aluminum on platinum in which the basal dimensions of the aluminum are those of the platinum support¹⁰). And there are several examples of materials that orient with a NaCl structure when grown on halide substrates^{11,12}). Hence, if a film of regular internal structure is required (the alternative being an amorphous deposit), it is important to select a substrate with crystal structure and cell size comparable to that of the sputtered material.

In those cases where a compound has more desirable mechanical or chemical properties than the elementally pure film and deposition by vacuum evaporation is difficult, reactive sputtering may be employed. By maintaining a suitable partial pressure of the reactant gas, oxides may be formed through in-flight or surface-catalyzed reactions of the metal vapor with oxygen, nitrides with nitrogen, carbides with methane or acetylene, and halides — care must be taken here to protect the sputter system, detector and pump — with the appropriate halogen gas¹³). Examples of metal-oxide targets that are difficult to prepare by standard evaporation techniques because of high melting points or susceptibility to decomposition include ThO_2 , SnO_2 , TiO_2 , CdO , PbO , and Al_2O_3 . In these cases, a suitable target may be made by reactive sputtering in an argon/oxygen atmosphere.

INCLUDED SPUTTERING GAS

A potential disadvantage to the fabrication of thin films by sputter deposition is the inclusion of gas in the sputtered material. The incorporated gas may alter the mechanical, chemical, and electrical properties of the film and, regarding nuclear applications, introduce

contaminant products in the nuclear-reaction spectrum.*

To determine the extent of inclusion of the argon discharge gas, foils were subjected to neutron-activation analysis. Several aluminum substrate foils onto which molybdenum had been sputtered were irradiated at the TRIGA Mark I Reactor Facility with a neutron flux of 3×10^{13} neutrons/cm²-s.

⁹⁹Mo and ⁴¹Ar, neutron-capture products of ⁹⁸Mo and ⁴⁰Ar, respectively, were studied by spectrum multiscaling with a shielded Ge(Li) gamma-ray detector and 4000-channel analyzer. Argon was found to be in the molybdenum host matrix with an abundance of from 3 to 7 atomic percent. Gas content was smaller when the discharge was sustained at relatively lower voltages with higher pressures.

Because high-energy neutral gas atoms (produced in charge-exchange interactions: $G_1^+ + G_2^0 \rightarrow G_1^0 + G_2^+$) are primarily responsible for film contamination⁸), limiting their energy spectrum through reduction of the cathode-fall potential difference and through thermalizing collisions with other atoms will decrease the extent of gas incorporation. Hence, at a system voltage and pressure of 2400 ± 100 V dc and 50 ± 10 mTorr, respectively, the Ar/Mo ratio was $7 \pm 1\%$; whereas, for a voltage of 325 ± 20 V dc and a pressure of 300 ± 10 mTorr, the ratio was found to be only $3 \pm 1\%$.

*In addition to the incorporation of the sputter-gas atoms, the possible presence of residual gases should be considered regarding the sputter deposition of the more reactive metals (for example, titanium, palladium, chromium, or molybdenum). Hydrogen and oxygen or other species from water vapor or hydrocarbons adsorbed on the system components or backstreaming from the vacuum pump may react with or embrittle the sputtered film¹⁴).

To substantially minimize the gas content of sputtered films, the substrate temperature may be allowed to approach the equilibrium value of about 300°C , depending on the plasma input power¹⁵). By thermally isolating the substrate with, say, ceramic standoffs, it will reach thermal equilibrium within minutes after ignition of the glow discharge. Films maintained at temperatures of 300°C or more show release of greater than 75% of the included gas^{16,17}) due to desorption following thermal migration of the implanted atoms.

CONCLUSIONS

Constructed from relatively inexpensive, off-the-shelf components, the sputtering system discussed above has been valuable in making thin films of aluminum, molybdenum (subject to oxidation), bismuth oxide, platinum, and gold and in reducing the thickness of foils of beryllium, aluminum, nickel, HAVAR, molybdenum, tantalum, platinum, and gold for work in nuclear science at this Laboratory. Some of these materials are commercially available as thin foils, and some may be prepared by other means. Sputtering is an alternative and a complement to other film-making techniques. It has documented advantages over other processes, and it can be economically attractive in providing foils of well-defined thickness.

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FIGURE CAPTIONS

- Fig. 1 Line drawing of the sputtering-electrode assembly.
- Fig. 2 Block diagram of circuitry associated with sputtering system.
- Fig. 3a Photograph of the cathode assembly.
- Fig. 3b Photograph of operating configuration. Alpha-particle detector is located beneath the substrate platform.

XBL 766 - 3009

Fig. 1

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Fig. 2

Fig. 3a

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Fig. 3b

XBB 766-4925

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