

## Deposition of Al/Cu Multilayer By Double Targets Cylindrical DC Magnetron Sputtering System

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### Abstract

A cylindrical direct current magnetron sputtering coater with two targets for deposition of multilayer thin films and cermet solar selective surfaces has been constructed. The substrate holder was able to rotate around the target for obtaining the uniform layer and separated multilayer phases. The Al/ Cu multilayer film was deposited on the glass substrate at the following conditions: Working gas = Pure argon, Working pressure = 1 Pa, Cathode current = 8 A and cathode voltage = -600 V .Microstructure of the film was investigated by X-Ray Diffraction and the scanning electron microscopy analyses. The elements profile was determined by glow discharge–optical emission spectroscopy analysis. During deposition, both targets with magnetron configuration were sputtered simultaneously by argon ions. A Plasma column on the targets surface was generated by a 290 G permanent magnet unit. Two DC power supply units with three phases input and maximum output of 12 A/1000V were used to deposit the multilayer thin films. A control phase system was used to adjust output voltage.

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## 1. Introduction

DC magnetron sputtering is the most usually practiced commercial form of sputtering. Magnetron sputtering technology can be used to deposit wide range of materials on any type of substrates including metals, ceramics and temperature sensitive plastic materials. It is

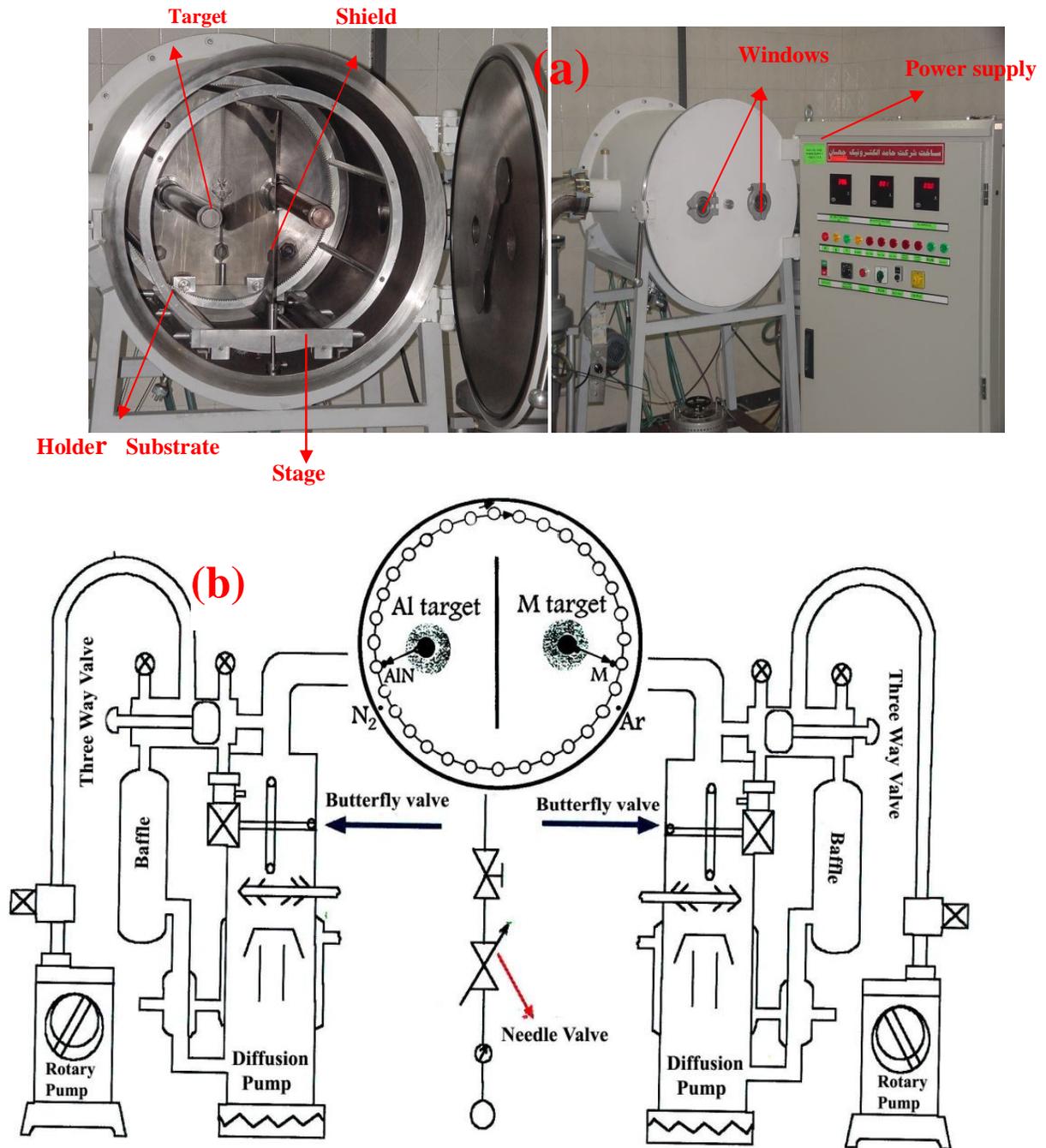
considered to be the best choice to deposit multilayer coatings. A composite coating usually consists of two or more phases combined either as different layers (multilayer) or as a homogeneous isotropic mixture of different phases (multiphase). The aim of such a coating is to combine the desired properties

from different components as well as creation of new properties generated by combination of suitable materials[1]. Many researches have done major efforts to improve performance of the deposition magnetron type system. T. T. Huang, et. al [2] were fabricated a cylindrical magnetron sputtering target assembly includes a head and a main body connecting to the head. The main body includes a target, a protecting element located partially around the target and an insulating assembly. The insulating assembly includes at least one insulating element which is fastened to the inner sidewall of the inner plate.

T. Nyberg et. al [3] were introduced a method for obtaining a reactive sputtering process with a reduced or eliminated hysteresis behavior by focusing the ion current onto a small area, a reduced erosion area which is in constant motion along the target to avoid melting of target material. This means that the current density is very high at the reduced erosion area while the average overall current density is significantly lower. The problem with arcing during reactive sputtering will be suppressed since the compound layer is effectively removed if the current density is sufficiently high which resulted in a substantial increase of the fraction of ionized sputtered species.

Multilayer coatings usually were used to enhance adhesion which can be attributed to formation of diffused interfaces. Another advantage of the composite coatings in multilayer design is that they can provide combination of properties such as high hardness, high toughness and low friction to satisfy various applications through selecting different coating partner systems [1]. In these cases, the multilayer deposition process is usually carried out by magnetron sputtering or reactive

magnetron sputtering technology. Multilayer conception provides the flexibilities to design coatings for specified applications. In designing the proper multilayer systems, the task can generally be divided into two areas: structural and functional designs. Multilayer structures can be tailored by change in geometry and morphology of the grains, grain size, growth orientations and thickness of the individual layer. In this way, with the change in the structural components ensure the coating to enhance toughness and hardness so as to produce more wear resistant coatings [1]. On the other hand, the functional design aspect includes selection of the material depends on the type of bonding (i.e., metallic, ionic, and covalent) and thermal, physical and mechanical properties of the material . Magnetron sputtering technology also has been used for deposition of solar selective absorbing coatings with improved solar performance, in particular, lower emittance and less environmental pollution than electrochemical methods. A cylindrical direct current magnetron sputtering coaters with two targets for deposition of thin multilayer of metals has been constructed. In order to produce cermet (metal-ceramic) solar coatings, two targets (Al and Cu) were run independently. The Cu target was used for deposition of a metal infrared reflection layer using DC sputtering in pure argon gas. While Al target was used for deposition of the ceramic layer in a gas mixture of argon and nitrogen. This device provides significant improvements in performance and permit discharges to be operated at pressures as low as  $10^{-4}$  Torr.



**Fig. 1.** (a) Double targets cylindrical DC magnetron sputtering system and (b) typical illustration of diagrammatic form of a cylindrical vacuum chamber and vacuum line

The process relies on a suitable ion bombardment of a target material by noble gas ions (normally  $\text{Ar}^+$ ) in glow discharge plasma. One can sequentially deposit a thin multi-layer stack by sputtering of the target atoms. The smooth and uniform interfaces can be formed if the substrate is rotated across the sputtering target instead of keeping stationary.

## 2. Methods and Results

To deposit the Al/Cu multilayer film on the substrate, a two target magnetron sputtering system was designed and successfully operated.

The vacuum chamber with 650 mm length and inner diameter of 550 mm is electrically grounded. The entire apparatus is mounted on the rails of a frame. In the side portion of vacuum container, an air suction port was provided which connected to a vacuum pump. The magnetron sputtering apparatus is shown in Fig.1(a).

The cylindrical cathode and a work piece were positioned within a cylindrical chamber which was evacuated to a pressure of about  $7.5 \times 10^{-4}$  Pa by diffusion pumps (See Fig. 1(b)). Two DC power supply units with three input phases and maximum output of 12/1000 A/V were used to deposit the multilayer thin films. The power supplies topology was specifically designed for usage in a plasma environment. It was optimized to be stable at high power of running process plasma. A control phase system was used to adjust output voltage. It is necessary to control automatically current and voltage when the surge current occurs [4-8]. The coater contains two targets with 550 mm length and 50 mm diameter that can be independently run. During deposition of thin layer both the targets were sputtered simultaneously by argon ions. They were separated by a screen to prevent their

contamination. The cathode was generally formed from electrically and thermally conductive metals such as copper, stainless steel, aluminum and so on. In order to prevent deviation or bending of magnetic lines, tubular shaped cathode was formed from a non-magnetic metal. A cylindrical target either formed or plated with the desired material for sputtering was seated to the chamber through an electrical isolator. The mounted target within the chamber was held by supporting means. A sputtering target assembly, including a target and target support member and a chamber backing plate to hold the backside of the target assembly was disposed at the central portion of the back side of the cylindrical chamber and extended into the vacuum chamber through the vacuum tight seals as shown in Fig. 2.

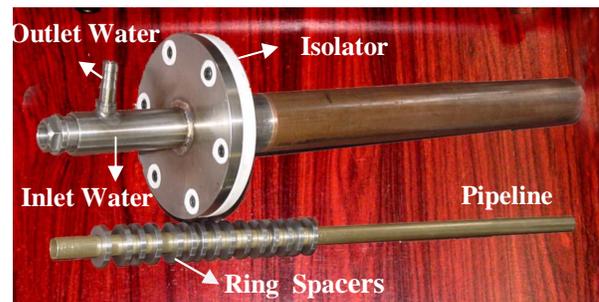


Fig. 2. Cylindrical cathode and cathode assembly.

A gas injection port in which a gas cylinder connected through a control valve was located at the bottom of the chamber. A sputtering working gas, preferably argon, was metered into the chamber along the targets through inlet jets in a stainless steel located at the bottom of the chamber. The gas injection flow rate was adjusted using a mass flowmeter. The metallic target was biased to a negative DC bias in the range of about 0-1000 V in order to attract positive ions of the working gas toward the target for sputtering the metal atoms.

A cylindrical magnet with a hole along the axis was provided within the target to provide an orthonormal electric and magnetic fields between the target and substrate. Multiple cylindrical magnets symmetrically oriented around the axis of cylinder for generating a toroidal (ring-shaped) magnetic fields tending to restrict charged particles to a volume around the cylindrical cathode. The cathode surface was located close to the magnet poles such that magnetic field lines penetrated to the cathode and formed a closed ring gap. In order to cooling the target during sputtering a pipeline was crossed within it. Multiple permanent magnets have the advantages of producing multi-toroidal volumes or particle "racetracks" to concentrate the charge particles which resulted in multiple erosion zones centered upon the center of the magnet. The magnets were disposed within the target to form a close ring of the magnetic force lines around the target [9-15]. The magnet package contains axially aligned ring-type permanent magnets mounted on a central actuating rod. The magnets were arranged with alternating polarity and joined with ferromagnetic ring spacers as a soft magnetic substance. It is preferable using the soft steel or iron for the soft magnetic material to provide a flux return path. The magnets were spaced from the inner cathode wall to allow the flow of coolant through the cathode in the annular space.

Cylindrical magnetrons have been designated in an arrangement whereby the magnetic field lines were bent to intersect with the cathode barrel. The created plasma by the trapped electrons essentially defines a cylindrical region around the target. Therefore, such systems are generally configured with several electron traps along the cathode. A trap for the primary electrons which restricts their motion both

radially and axially, causes to remain the electrons near the target surface until a large fraction of their energy has been expended in ionizing collisions [11,12]. In a cylindrical magnetron, the  $E \times B$  electron drift; perpendicular to both of the electric and magnetic fields, causes orbital motion of the electrons around the target. Unfortunately, the electrons tend to leak out or escape their orbits close to each end of the cylindrical target, resulting in lower ionization intensities and thus lower sputtering rates [15,16]. The bias voltage and magnetic field caused considerable amount of energy to be dissipated by the target and magnetron, thereby tending to heat the target and magnetron. It should be noted that heating of the magnetron and target above a designated processing temperature may adversely affect the performance of system by changing the sputtering rate or reducing uniform sputtering of the target. Additionally, excess heat may be shortening the useful life of the magnetron and target and caused mechanical features of it to wear prematurely out. Furthermore, it may be caused thermal expansion of components within the chamber, which can cause closely spaced components such as target and magnetron to physically interfere with each other. To alleviate this problem a pipeline introduced coolant within the cylindrical target. Coolant inlet and outlets were provided at either end of the cathode, such that a coolant fluid such as water was flowed in the annular space between the magnets and the inner of cathode wall. Coolant inlet pipes, suitably insulated, were connected to the coolant inlets which were sealed at the other end of each cathode through the O-rings.



Fig. 3. (a) Substrate holder and (b) its stage.

The work-piece holder was positioned between the cathode barrel and the side wall of chamber which made from non-magnetic material. During sputtering the substrates were accommodated on the holder which it can be rotated around the center axis of the chamber by a controlled variable speed of DC motor. The connection of this motor to the shaft caused to rotate the holder around the central axis. The holder was held on a stage. The holder and its stage are shown in Fig. 3. To move the holder horizontally, the stage was located on rollers [16-20]. To clarify, the operated system efficiency and performance, it has been compared with the similar works published recently in Table 1. To estimate performance of the system, Al-Cu multilayer film was deposited on the glass substrate [25-27]. The microstructural and morphology of the film was studied by X-ray diffraction (XRD) and scanning electron microscope (SEM), respectively.

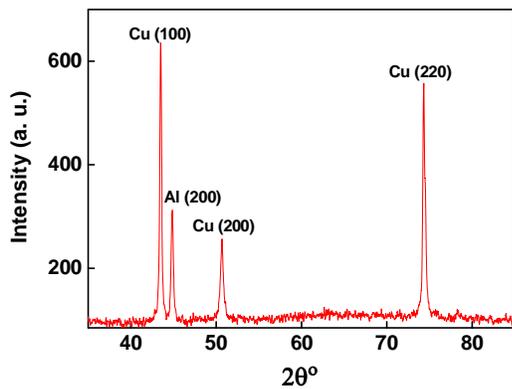
Table.1 Comparison of the home made magnetron sputtering systems.

Pressure (Torr)	Cooling System	Power supply	Substrate Temp (°C)	Sputterin gas	Target	Magnetic field (G)	Vaccum pump	Type	Reference
10 <sup>-5</sup> †	Water	DC	300	Ar	Cylindrical	244	Rotary, Diffiusion	Semi-industrial	Our work
9×10 <sup>-9</sup>	Water	DC	Room temperature	Ar	Planar	---	Rotary, Diffiusion	Researching	[21]
10 <sup>-7</sup> -10 <sup>-9</sup>	Water	DC, RF	850	Ar	Cylindrical	----	Rotary, Turbo	Researching	[22]
10 <sup>-7</sup>	Water	DC	Room temperature	Kr	Cylindrical	2000	Rotary, Turbo	Researching	[23]
10 <sup>-6</sup>	Water	DC	600	Ar	Planar	----	Rotary, Turbo	Researching	[24]

† Due to Chamber volume(300 Lit) & pumping speed of diffiusion pump (280 Lit/s).

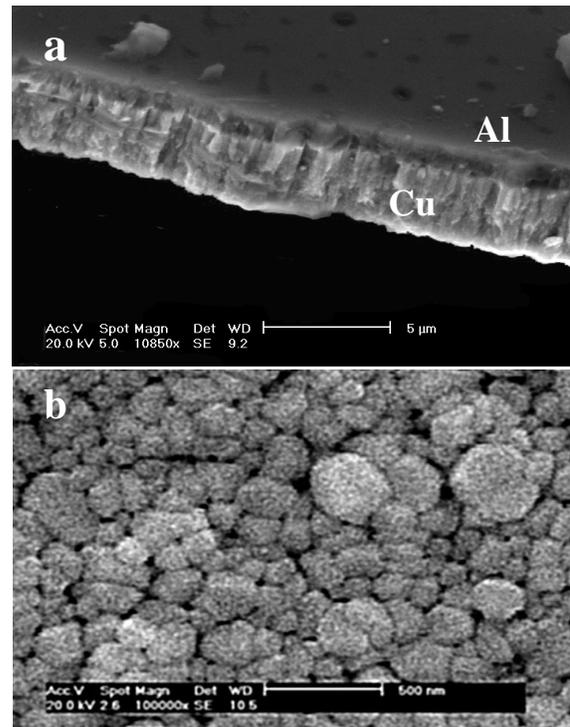
Fig. 4 presents XRD pattern of a Cu/Al/glass system prepared at 250 °C. It shows an intense fcc-Al (200) peak at 2θ=44.82° (from 00-001-

1176 JCPDS number) and one intense fcc-Cu (100) peak at  $2\theta=43.3^\circ$  as well as much weaker Cu (200) and (220) peaks at correspond positions of  $2\theta=50.6^\circ$  and  $2\theta=74.3^\circ$  (from 00-001-1242 JCPDS number), respectively. As can be seen Al and Cu were grown on two separate phases which can be considered as a proof for formation of multilayer structure of the film.



**Fig. 4.** X-ray diffraction patterns of Al/Cu multilayer film at 250 °C.

Fig. 5(a) shows cross-sectional SEM micrograph of Al/Cu/glass multilayer coating with a columnar growth structure on the substrate. Two distinct Al and Cu layers can clearly observe. Top view SEM micrograph indicates non-uniform distribution of the grain size on the film surface (Fig. 5b). The average grain size of the upper layer was estimated to be 113 nm which is in agreement with crystallite size obtained from XRD pattern by Sherrer relation. Fractal concept was widely used to characterize the complexity of the films surfaces. It correlated with the growth modes of the film and surface roughness [28]. Fractal dimension (FD) parameter can be calculated from topographic images from the film surface.



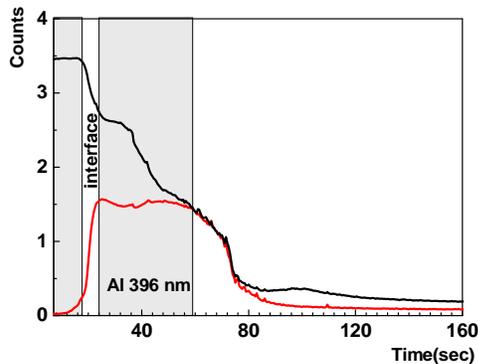
**Fig. 5.** (a) The cross sectional and (b) top view SEM micrograph of Al-Cu multilayer film.

According to difference between gray levels of the SEM images, surface characterization is possible. The histogram of the heights on the film surface was obtained using Imagej software. They exhibit a sharp peak that can be well fitted using a Gaussian function. FWHM of the Gaussian distribution, average grain size and fractal dimension are presented in Table 2. They describe topography and complexity of the film surface. As shown, a rough surface with high complexity is formed. The chemical composition-depth profiles of the Al/Cu multilayer were evaluated by glow discharge optical emission spectroscopy (GD-OES) for deposited layers (Fig. 6).

**Table 2.** Parameters calculated from SEM image of Al-Cu multilayer.

FWHM	FD	Grain size (nm)
78.53	1.87	113

As can be seen, depth profile of Al and Cu elements is measured as 327 nm and 396 nm, respectively which decrease over the time.



**Fig. 6.** Glow discharge-optical emission spectroscopy (GDOES) of Al-Cu multilayer film.

## Conclusions

A novel direct current magnetron sputtering coating system with two targets for depositing of multilayer thin films for cermet solar selective surfaces has been designed and operated. The substrate holder can rotate around the target for obtaining the multilayer and separated multilayer phases. This system provides significant improvement in coating performance. The vacuum chamber was 650 mm length with an inner diameter 550 mm. The coater contained two cylindrical targets 550 mm long and 550 mm diameter that can be independently run. They were protected by a screen to prevent their erosion. The Al/Cu multilayer film was deposited on the glass substrate. The X-Ray Diffraction and the scanning electron microscopy were used to identify the two layers and study the

film morphology, respectively. The elements profile was determined by glow discharge-optical emission spectroscopy.

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