Aluminum films deposition by magnetron sputtering systems: Influence of target state and pulsing unit

D V Sidelev¹, A V Yuryeva¹, V P Krivobokov¹, A S Shabunin¹, M S Syrtanov¹, Z Koishybayeva¹
¹Institute of Physics and Technology, Tomsk Polytechnic University, Tomsk 634050, Russia

Abstract. This article reports on technological possibilities of magnetron sputtering systems with solid-state and liquid-phase targets to deposition of aluminum films and its structure. The comparison of deposition rates of magnetron sputtering systems with direct current (DC), mid-frequency (MF) and high power pulsed (HiPIMS) supplies is shown. The optical emission spectroscopy indicates a high component of target material ions in discharge gap only to HiPIMS technique. Al films are a (111)-line oriented in DC and MF power supply cases, for high power pulsed unit – aluminum films also have intense (220)-line. The dependence of grain sizes and sputtering technique parameters is obtained.

1. Introduction

Al films have been widely employed as a reflective layer with high electrical conduction. Such coatings are interested to space applications as being a reflector element of satellite [1]. There is required a thick Al layer (1-3 µm) with radio-frequency transparent protective upper-layer (SiO₂ or Al₂O₃). Now, ion-plasma techniques are demand to thin films deposition [2-5]. These technologies are enabled to required thickness uniformity on large-scale substrates, structure homogeneity, high purity and adhesion, low heat flow on substrate. Thus, magnetron sputtering techniques are used to deposit of Al-based functional coatings on low-melting and carbon composite materials. Above, our colleagues made technology of thermal-control coatings deposition, based on ion-plasma techniques instead of resistive evaporation [6]. Such way has been helped on increase of output of technological process and part of nondefective units (up to 90%).

Today, the actual tasks in magnetron sputtering techniques are to increase a deposition rate(a) and adhesion of growing coatings to substrate(b), deposit films with predictable properties(c). Therefore, HiPIMS and liquid-phase target magnetrons are attractive. The first technique has a low deposition rate [7], but form a short-term (30-200 µs) high-current discharge with plasma density 10¹³ cm⁻³, for comparing DC and MF magnetrons have only 10¹¹ cm⁻³. The last investigations of HiPIMS techniques are indicated on film densification [8], increase adhesion [9] and others changing in film properties. The unique construction of the second magnetron type has a special pot to melting target [10] and evaporation initiation. Thereby, the significantly increase of target erosion due to combined mechanism behaviour was. In this case, the deposition rate of metal coatings can be enhanced to 10²-10³ nm/s. Moreover, the so-called “self-sputtering” process can initiates in liquid-phase target technique and magnetron discharge changes into a self-sustained mode. For this case, plasma discharge operates on metal vapor at 7×10⁻³-2×10⁻² Pa. These technologies are primarily interested for ultra-high deposition rates and film purity. Such magnetron sputtering techniques can be beneficial to Al films deposition for...
space applications. In this study the comparing of technological possibilities of different magnetron sputtering systems to Al films deposition and its structure investigation were done.

2. Experimental

Aluminum films were deposited in an ion-plasma vacuum installation by magnetron sputtering systems with Al target (99.995%). The base pressure was $5 \times 10^{-3}$ Pa. Here, magnetrons with solid-state disk (dia. 90 mm) and liquid ring-shaped (inner dia. - 54 mm, outer dia. - 104 mm) targets were used. Three different power supply have been chosen. The pulsed DC-power supply was used to liquid-phase target magnetron. MF (1-100 kHz) and HiPIMS (0.1-15 kHz) pulsing units have been connected with solid-state target magnetron. The discharge parameters were measured by oscilloscope Tektronix 2022 B. All depositions were performed in the power stabilization on mode. For substrates, $30 \times 30$ mm² borosilicate glasses were used. We use a specific deposition rate ($v_s$), which refers to thickness deposited per time and power of pulsing unit, in nm/(kW×s).

Table 1. Deposition modes of Al films

| №  | Target state | $W_{av}$, kW | Power unit parameter | $P$, Pa | $h$, µm | $v_s$, nm/(kW×s) |
|----|-------------|--------------|---------------------|--------|--------|------------------|
| s1 | solid       | 2.5          | 100 8 2             | 0.18   | 0.5    | 0.49             |
| s2 | solid       | 2.5          | 1 800 200           | 0.5    | 0.5    | 0.45             |
| s3 | solid       | 2.5          | 1 140 860           | 0.51   | 0.49   | 0.185            |
| s4 | solid       | 2.5          | 1 140 860           | 0.51   | 0.49   | 0.185            |
| s5 | solid       | 2.5          | 1 140 860           | 0.51   | 0.49   | 0.185            |
| s6 | solid       | 2.5          | 1 140 860           | 0.51   | 0.49   | 0.185            |
| l1 | liquid      | 3            | 10²(*) 1.2          | 6      | 0.4    | 10               |
| l2 | liquid      | 3.2          | 0.23               | 0.42   | 7.7    |
| l3 | liquid      | 3            | 10²(*) 1.2          | 6      | 0.4    | 10               |
| l4 | liquid      | 3            | 10²(*) 1.2          | 6      | 0.4    | 10               |

Note: $W_{av}$ – average power; $v$ – pulse frequency; $t_0$ – operating pulse; $t_p$ – pause between pulses; $P$ – pressure; $h$ – film thickness; * - self-sputtering mode.

The effect of power supply on the magnetron discharge properties was studied by optical emission spectroscopy. The double-channel (225-920 nm) fiber-optic spectrometer AvaSpec was used. The integration time was 5 ms, the number of measurements for averaging – 100, focal distance – 300 mm. We use NIST atomic spectra database (ver. 5.3) for identification spectral lines of plasma discharge.

The film thickness was analyzed by spherical microsection method (Calotest CAT-S0000, CSEM). XRD measurements of Al films were performed using a Shimadzu XRD-7000S with accelerating tube Cu-Kα (40 kV and 30 mA) in 2θ from 30º to 90º with exposure step 0.03º. The scanning angle was 1º.

3. Results and discussion

A pulsed DC and MF magnetron discharges strongly depend on the operating mode. There are three alternant processes: plasma build-up, stationary plasma and decaying plasma process [11]. Any recorded operating pulse parameters by oscilloscope are illustrated it. Figure 1 corresponds to specific deposition rates on pulse frequency to DC and MF solid-state target magnetron system. The stationary discharge mode is the longest-term at 1 kHz and current pulse has a practically rectangular shape. The operating voltage is an identical to DC mode, so $v_s$ (for 1 kHz) is less to DC mode due to decaying plasma phase. The change of pulse frequency from 1 to 100 kHz at constant duty cycle results in decrease of $t_p$. So, there is an increasing of residual ionization from 0 to 3.36 A, deposition converts from pulse mode to “quasi-stationary” at high pulse frequency (plasma build-up phase is shorten). The specific deposition rate is higher to 60-100 kHz than for DC mode due to increase a discharge voltage. The low $v_s$ in1-40 kHz is correlated with non-optimal time-ratio of plasma phases and discharge voltage.
Figure 1. The specific deposition rate and residual ionization on pulse frequency of MF magnetron. The label (*) is aligned to \( v_s \) of DC technique with solid target.

The dependences of \( v_s \) on pulse frequency and duty cycle of HiPIMS technique are shown in Figure 2. The specific deposition rate is linearly decreased in 1-7 kHz. The increase of pulse frequency is caused to change of current pulse shape from rectangular to triangular. Firstly, the current increased and saturated to 24 A (for 1 kHz), then the discharge current stabilized. For 7-15 kHz, the pulse current increased at permanent average power 2.5 kW (33 A for 15 kHz).

The decrease of duty cycle is caused in fall of \( v_s \). It is resulted in decrease of stationary plasma phase and increase pulse current. In this case, the self-sputtering mode is affected on productivity of magnetron sputtering system [12]. The difference between \( v_s \) for HiPIMS and MF discharge is non-stealthy.

Figure 2. The specific deposition rate on pulse frequency (a) and duty cycle (b) of HiPIMS technique.

The main advantage of liquid-target magnetron is the high deposition rate. The specific deposition rate is 42-163 times higher than for high power impulse magnetron and 14-50 times - MF sputtering. The main mechanism of increase \( v_s \) is an additional evaporation of liquid-state target to sputtering [6].

Figure 3.a corresponds to optical emission spectra of magnetron discharge. The DC and MF sputtering systems have practically equal spectral patterns. It is a high content of Ar and Al atoms and relatively low concentration of Ar\(^+\) and Al\(^+\) ions. The emission spectra of HiPIMS technique have significant differences. There are increases of concentration of ions components (Ar\(^+\), Al\(^+\)) and falling of neutral atoms intensity (Ar). Thus, the dense plasma forms in the HiPIMS case. Moreover, the decrease of Al intensity (394.4 nm) confirms of fall \( v_s \) comparing to the MF discharge.

Figure 3.b corresponds to emission lines of Ar atoms (842.47, 912.29 nm), Ar\(^+\) (358.84, 465.79, 704.21 nm) and Al\(^+\) (281.69, 623.18 nm) ions to the high power pulsed magnetron discharge. The obtained results have a good agreement with data of deposition rates and shapes of current pulses. The pulse time has a predominant influence of plasma composition. The generation mechanism of Al\(^+\) ions
is an ionization of Ar atoms\(^{(a)}\), sputtering of Al target\(^{(b)}\) and ionization of Al atoms\(^{(c)}\). The increase of \(\text{Al}^+\) ions is inherent to long-time pulses and initiated by drop of pulse frequency at constant duty cycle. It results in effect of ion bombardment to substrate. On the contrary, the positive ions of aluminium can chance to accelerate to cathode and take a part in sputtering of Al target. There is a “self-sputtering” and its probability increases with \(t_0\).

![Image](https://via.placeholder.com/150)

**Figure 3.** (a) The optical emission spectra of magnetron discharge: 1 – DC mode; 2 – MF, 1 kHz; 3 – MF, 40 kHz; 4 – MF, 100 kHz; 5 – HiPIMS, 1 kHz; 6 – HiPIMS, 7 kHz; 7 – HiPIMS, 15 kHz. (b) Emission lines of Ar atoms, \(\text{Ar}^+\) and \(\text{Al}^+\) ions on pulse frequency of HiPIMS discharge.

The diffraction patterns are shown in Figure 4. The textures of DC and MF coatings are equal. There are high-intensive (111) and low-intensive (200), (220), (311) crystallographic directions. The grain sizes are 20-24 nm calculated by Debye-Scherrer formula.

![Image](https://via.placeholder.com/150)

**Figure 4.** XRD structure of Al films: a – solid-state target magnetron; b - liquid target magnetron.

The HiPIMS coatings have more intensive (220) line. The decrease of pulse frequency is resulted in forming structure with dominant (111) and (220) intensities. Moreover, narrowing of (111) lines to 0.4-0.5 \(\mu\)m-thick films are indicated to form coarse-grained coatings. It is good illustrated to Al films deposited by HiPIMS and liquid-target techniques. For high power pulsed magnetron sputtering system,
grain sizes are more 26 nm, for liquid target magnetron – 53 nm and higher. In the first case, it can be resulted in forming denser structure due to the higher concentration of Al\(^+\) ions in deposition flow. For liquid target technique, the high-intensive evaporation is caused in grain growth.

4. Conclusions
The solid-state and liquid target magnetron sputtering systems with different pulsing units were used to Al film deposition. The comparing of technological possibilities of deposition techniques shows on a variance of specific deposition rates and plasma discharge composition. The liquid target magnetron has a more productive mode (6.25-24.5 nm/(kW\(\times\)s)) and forms coatings with high grain sizes (higher 53 nm) due to intensive evaporation of liquid target material. The using of HiPIMS technique is caused in the increase of ion component Al\(^+\) in deposition flow and grains growth of Al coatings to 26 nm and higher.

Acknowledgments
This investigation was supported by Russian Science Foundation (project № 15-19-00026).

References
[1] Asainov O H, Bainov D D, Krivobokov V P, Romanenko S E, Chernyatina A A 2011 Russian Physics Journal 54 158
[2] Erofeev E V, Kagadei V A, Kazimirov A I, Fedin I V 2015 Proc. Int. Sib. Conf. on Control and Communications SIBCON (Omsk) 7147054 (St. Petersburg: Russia / Institute of Electrical and Electronics Engineers)
[3] Kashkarov E B, Nikitenkov N N, Syrtanov M S, Babihina M N 2016 IOP Conf. Series: Materials Science and Engineering 110 012046 1
[4] Nikitenkov N N, Sutygina A N, Shulepov I A, Sivin D O, Kashkarov E B 2015 IOP Conference Series: Materials Science and Engineering 81 012018 1
[5] Yurjev Y, Sidelev D 2013 Journal of Physics: Conference Series 479 012018 1
[6] Kashkarov E B, Nikitenkov N N, Syrtanov M S, Sutygina A N, Shulepov I A, Lider A M 2016 Applied Surface Science 370 142
[7] Asainov O H, Bainov D D, Krivobokov V P, Mihailov M N, Pachenko O V, Ydakov C V, Chernyatina A A 2011 Russian Physics Journal 54 154
[8] Anders A 2010 Journal of Vacuum Science & Technology A 28 2010
[9] Samuelsson M, Lundin D, Jensen J, Raadu M A, Gudmundsson J T, Helmersson U 2010 Surface & Coatings Technology 205 591
[10] Bandorf R, Waschke S, Carreri F C, Vergöhl M, Grundmeier G, Bräuer G 2015 Surface & Coatings Technology, in press.
[11] Bleykher G A, Krivobokov V P, Yurjeva A V, Sadykova I 2016 Vacuum 124 11
[12] Musil J, Lestina J, Vcelek J, Tolg T 2001 Journal of Vacuum Science and Technology A 19 420
[13] Sarakinos K, Alami J, Konstantinidis S 2010 Surface & Coatings Technology 204 1661