JVST A
Journal of Vacuum Science & Technology A

Reactive pulsed magnetron sputtering process for alumina films

Cite as: Journal of Vacuum Science & Technology A **18**, 2890 (2000); https://doi.org/10.1116/1.1319679
Submitted: 17 March 2000 . Accepted: 28 August 2000 . Published Online: 10 November 2000

P. J. Kelly, P. S. Henderson, R. D. Arnell, G. A. Roche, and D. Carter

Reactive pulsed magnetron sputtering process for alumina films

P. J. Kelly,[a] P. S. Henderson, and R. D. Arnell
Centre for Advanced Materials and Surface Engineering, University of Salford, Salford, M5 4WT, United Kingdom

G. A. Roche and D. Carter
Advanced Energy Industries Inc., Fort Collins, Colorado 80525

(Received 17 March 2000; accepted 28 August 2000)

The pulsed magnetron sputtering (PMS) process is now among the leading techniques for the deposition of oxide films. In particular, the use of pulsed dc power has transformed the deposition of dielectric materials, such as alumina. The periodic target voltage reversals during the PMS process effectively discharge poisoned regions on the target. This significantly reduces the occurrence of arc events at the target and stabilizes the deposition process. Many researchers have now shown that pulsed dc reactive magnetron sputtering can be routinely used to produce fully dense, defect-free oxide films. Despite the success of the PMS process, few detailed studies have been carried out on the role played by parameters such as pulse frequency, duty cycle, and reverse voltage in the deposition process. In this study, therefore, alumina films were deposited by reactive pulsed dc magnetron sputtering. Operating conditions were systematically varied and the deposition process monitored throughout. The aim was to investigate the influence of the pulse parameters on the deposition process, and the interrelationships between the occurrence of arc events and the parameters chosen. As a result of this investigation, optimum conditions for the production of high-quality alumina films under hard arc-free conditions were also identified. [S0734-2101(00)04806-5]

I. INTRODUCTION

Since the initial development work in the early 1990s,[1–4] the pulsed magnetron sputtering (PMS) process has become established as one of the leading techniques for the deposition of oxide films. In particular, the use of pulsed dc power has transformed the deposition of dielectric materials, such as alumina.[1–3,5–9] The process itself has been well described in various review articles,[3,6,8–14] and no repetition is required here. It is sufficient to state that pulsed dc reactive magnetron sputtering offers significant advantages over conventional, continuous dc processing.[14] If the magnetron discharge is pulsed in the bipolar mode (see Fig. 1) at frequencies, usually, in the range 10–200 kHz, the periodic target voltage reversals effectively discharge poisoned regions on the target. This significantly reduces the occurrence of arc events at the target and stabilizes the deposition process. Many researchers have now shown that pulsed dc reactive magnetron sputtering can be routinely used to produce fully dense, defect-free oxide films. All stoichiometries are available,[5,6,8] arc events are suppressed,[1–3,6–9,15–17] deposition rates can approach those obtained for metallic films,[2,3,7,15,16] and in dual-cathode systems, very long-term (>300 h) process stability is attainable.[18,19] As a consequence, very significant improvements have been observed in the structure,[5,7,8] hardness,[7,8] and optical properties[6,13] of PMS alumina films, compared to dc sputtered films.

The target voltage wave form during asymmetric bipolar pulsed dc sputtering is shown schematically in Fig. 1. Referring to Fig. 1, the critical parameters which make up the wave form are the pulse frequency, duty factor, and reverse voltage. Duty factor is the relative proportion of the pulse cycle made up of the ''pulse-on'' period, when the target voltage is negative and sputtering is occurring. The reverse voltage is the nominal positive target voltage achieved during the ''pulse-off'' period, often expressed as a percentage of the mean-negative voltage during the pulse-on period. The schematic wave form in Fig. 1 shows a pulse frequency of 100 kHz, with a duty factor of 80%, and the reverse voltage set at 20% of the pulse-on voltage. In practice, this ''square'' wave form is not achieved due to the inherent characteristics of the plasma and the power delivery system, with both positive and negative voltage overshoots being observed.[20] These artifacts can be clearly seen in Fig. 2, an oscilloscope trace of the target voltage wave form obtained when actually operating under the conditions defined previously.

Reference has already been made to the many examples in the literature of the success of the PMS process. However, as yet, few detailed studies have been published on the role played by the pulse parameters in the deposition process. Belkind, Freilich, and Scholl,[9,10] derived an expression showing that the critical pulse frequency for arc-free operation depends on the discharge current and the pulse-off time. Although not explicitly stated, their study indicates that, for a given discharge current, the duty factor is actually the most critical parameter in establishing arc-free conditions. Also, these studies did not consider time-dependent effects, since arc counting was only carried out for 3 min per run. In situations where, during each pulse-off cycle, the parameters selected only partially discharge the poisoned regions on the target, a residual charge will accumulate until, eventually,

[a]Electronic mail: p.kelly@salford.ac.uk

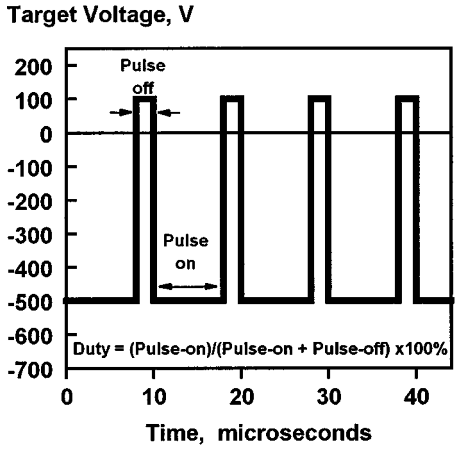

FIG. 1. Schematic representation of the target voltage wave form during asymmetric bipolar pulsed sputtering (pulse frequency=100 kHz, reverse time=2 μs, duty=80%, and reverse voltage=20%).

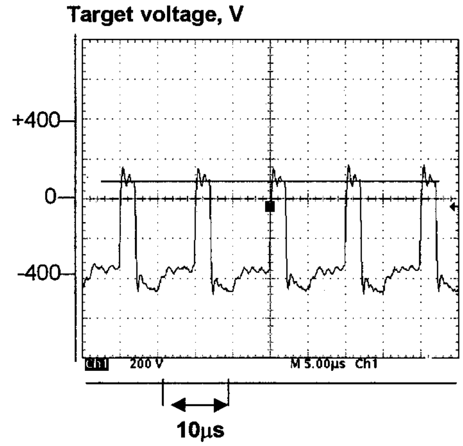

FIG. 2. Oscilloscope trace of the target voltage wave form when operating in asymmetric bipolar pulsed mode at 100 kHz (80% duty and 20% reverse voltage).

arcing occurs. Thus, conditions which appear to prevent arcing at the beginning of a deposition run can prove ineffective as the run progresses.

In this study, alumina films were deposited by reactive pulsed dc magnetron sputtering. Operating conditions were systematically varied and the deposition process monitored throughout. The aim was to investigate the influence of the pulse parameters, such as pulse frequency, duty factor, and reverse voltage, and the interrelationships between the occurrence of arc events and the parameters chosen. As a result of this investigation, optimum conditions for the production of high-quality alumina films under hard arc-free conditions were also identified.

II. EXPERIMENT

The commercial interest generated by the PMS process has led to the development of new power delivery systems. These include ac supplies, single- and dual-channel pulsed dc supplies, and pulse units which can be connected in series with the output from standard dc magnetron drivers. This article concentrates on the use of this latter type of system, in which the magnetron discharge could be pulsed over the frequency range 1–100 kHz. Parallel studies are also being made of the latest generation of pulsed dc supply which extends the maximum pulse frequency up to 350 kHz.[21,22]

The dc power supplies used in this study were the Advanced Energy MDX and Pinnacle magnetron drivers. These power supplies were used in conjunction with the Advanced Energy Sparc-le V pulse unit. The Sparc-le V unit allows the pulse parameters to be varied over the following ranges; frequency: 1–100 kHz, reverse time: 1–10 μs, and reverse voltage: 10%–20%. The dc supplies were operated in current regulation mode.

The Sparc-le V unit allows both hard arc and microarc events to be monitored. Hard arcs are generally considered to be a discharge which takes place between a region on the cathode and an earthed surface, whereas microarcs are dis-

arcs can normally be tolerated, hard arc events are extremely detrimental to the deposition process.[3,8] Thus, in this study only the incidence of hard arcs was monitored.

The work performed here was carried out in a Teer Coatings Ltd. UDP 450 closed-field unbalanced magnetron sputtering rig, which has been described in detail elsewhere.[7,8] Alumina films were deposited by reactive unbalanced magnetron sputtering from a 99.5% pure Al target. In all cases the base pressure was $<2\times10^{-5}$ mbar, the argon flow rate was adjusted to give a chamber pressure of 2×10^{-3} mbar prior to deposition, and the target current was set to 6 A. The target was precleaned with the substrates shuttered, but no sputter cleaning of the substrates themselves was carried out. In fact, the substrate holder was allowed to float electrically throughout. The flow of reactive gas was controlled by an optical emissions monitoring (OEM) system tuned to the 396 nm line in the Al emission spectrum. An OEM turn-down signal of 25% was used for all depositions, i.e., reactive gas was allowed into the chamber until the OEM signal had fallen to 25% of the initial 100% metal signal. A feedback loop then maintained the OEM signal at this value for the duration of the deposition run, which was typically 90 min. Previous experience had shown that such conditions would produce stoichiometric Al_2O_3 films.[8]

Figure 3 shows the characteristic hysteresis behavior of this system as the oxygen flow rate is varied. As the oxygen flow is increased initially, the target voltage rises slightly. Operating in this ''metallic'' regime could result in the formation of a substoichiometric aluminum oxide film. At a flow rate of approximately 13 sccm of oxygen, the target poisons rapidly and the negative target voltage falls from 395 to 250 V. The target then remains poisoned until the O_2 flow rate is reduced to <4 sccm. Operating in the ''poisoned'' regime would produce stoichiometric films, but at very much reduced deposition rates. The OEM system allows control to be maintained at any point on the hysteresis curve. Figure 4 shows the relationship between target voltage and the OEM setting, expressed as a percentage of the 100% metal signal.

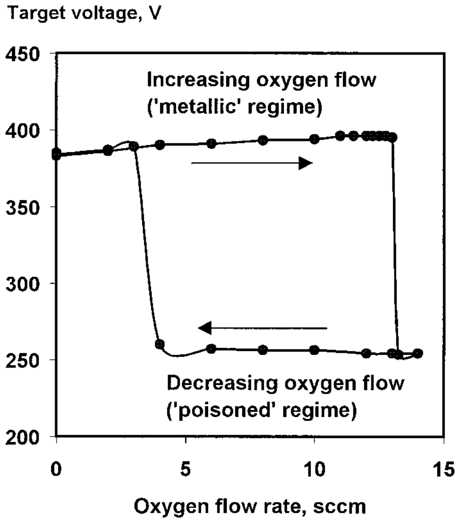

FIG. 3. Hysteresis behavior displayed during reactive sputtering of alumina.

maintains the target between the metallic and poisoned regimes in a ''partially poisoned'' mode. This allows stoichiometric Al_2O_3 films to be deposited at acceptable rates.

The first stage of this investigation was to deposit a series of alumina films under systematically varied conditions using the Pinnacle/Sparc-le V combination referred to above. For each run, the total number of hard arcs detected by the Sparc-le V was recorded. The film properties were then investigated, and the effectiveness of the deposition conditions at arc suppression was considered. The Taguchi method[23] was used to design this experiment. This method utilizes fractional factorial arrays which are designed to optimize the amount of information obtained from a limited number of experiments, and, as such, it is a very efficient experimental technique. The Taguchi L9 array was selected, which allows up to four factors to be varied at three levels, although only three factors were actually used. The factors chosen were

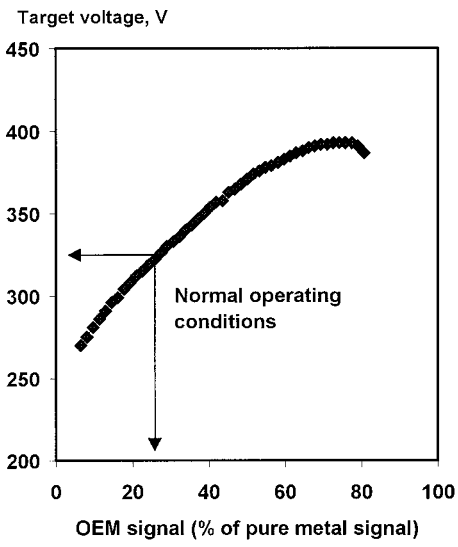

FIG. 4. Relationship between optical emission (OEM) signal and target volt-

TABLE I. Experimental Taguchi L9 array for the investigation of alumina films.

Run No.	Pulse frequency (kHz)	Reverse time (μs)	Reverse voltage (%)
1	20	1	10
2	20	5	15
3	20	10	20
4	35	1	15
5	35	5	20
6	35	10	10
7	50	1	20
8	50	5	10
9	50	10	15

pulse frequency (at levels 20, 35, and 50 kHz), reverse time (1, 5, and 10 μs), and reverse voltage (10%, 15%, and 20% of the nominal sputtering voltage). This range of frequencies was chosen because the Sparc-le V limits the maximum reverse time which can be selected at frequencies greater than 50 kHz. Higher frequencies were explored in a second array. The initial experimental array is summarized in Table I.

The alumina films were deposited onto precleaned glass substrates which were subsequently sectioned for analytical purposes. The coating structures were examined by scanning electron microscopy (SEM), with the thickness of each coating being measured from fracture section micrographs. Deposition rates were then calculated from these measurements. The composition of the coatings was determined using a JEOL JXA-50A microanalyzer equipped with WDAX. A high-purity aluminum standard was used in the analysis, with oxygen content being determined by difference. X-ray analyses were carried out using a Philips system, operating in θ–2θ mode (Cu $K\alpha$ radiation), and the resistivity of the coatings was measured using a four-point probe.

Following this, a second array of experiments was carried out. In this case, coatings were deposited over an extended range of pulse frequencies, up to 100 kHz. Also, the MDX magnetron driver was used as the dc supply to allow comparison with the Pinnacle unit. Deposition runs were repeated under, otherwise, identical conditions, but at different levels of duty factor. Care was taken between runs to sputter clean the target, such that all runs started with the target in a similar condition. Run times were varied to ensure that the total pulse-on time was consistent, i.e., the total sputtering time was constant. The reverse voltage was fixed at 20% of the nominal sputtering voltage. The number of hard arcs displayed by the Sparc-le V was recorded at regular intervals, both to monitor the onset of arcing, and to give the total cumulative number of arc events for each set of conditions. The coating structures and properties were investigated as for the preceding array.

III. RESULTS

The deposition rates and total number of hard arcs recorded during each of the Taguchi array runs are listed in Table II. The deposition rates have been normalized to target

TABLE II. Taguchi L9 array data table.

Run No.	Duty factor, %	No. of hard arcs recorded	Coating thickness (μm)	Normalized dep'n rate (nm/min/A)
1	98	>10 000	4.5	10.0
2	90	1823	3.0	9.3
3	80	5	1.4	4.6
4	96.5	>10 000	2.0	6.5
5	82.5	492	4.0	8.8
6	65	0	0.75	2.3
7	95	>10 000	3.75	11.6
8	75	2754	1.4	4.6
9	50	0	1.8	4.0

number of arcs which can be displayed by the counter on the Sparc-le V is 10 000. Where this value was reached before the end of a run, a value of >10 000 has been inserted in Table II. Also listed in Table II are the duty factors for each run, arising from the array settings of pulse frequency and reverse time. Statistical analyses were carried out on these data using a software package from the American Supplier Institute, entitled ANOVA-TM. This package was used to compute the level averages using deposition rate and number of hard arcs as response variables, i.e., to compute the average response of each variable at each level of each factor. The results of these analyses are shown graphically in Figs. 5 and 7, respectively. It appears from Fig. 5 that reverse time and reverse voltage both have significant, but opposite influences, on deposition rate. In the case of reverse time, this is simply because, as this factor is increased, so the pulse-off time becomes a greater proportion of the total pulse cycle, i.e., the duty factor is reduced and sputtering takes place for a lesser proportion of each cycle. This is illustrated in Fig. 6, which shows the positive correlation between the duty factor and normalized deposition rate (correlation coefficient, r =0.77). Thus, it could be argued that, of the variables investigated, reverse voltage actually has the most significant influence on deposition rate. As reverse voltage is increased from 10% to 20%, the level average for the normalized deposition rate increases from 5.6 to 8.3 nm/min/A, a factor of approximately 1.5 times.

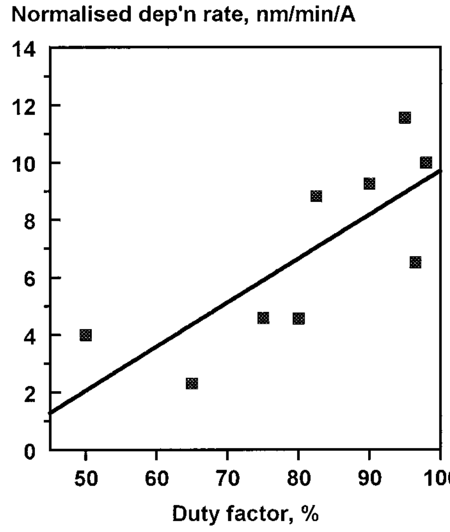

FIG. 6. Relationship between the duty factor and normalized deposition rate for reactive pulsed sputtered alumina films.

The Taguchi analysis using the total number of hard arcs detected as the response variable is shown in Fig. 7. Rather surprisingly, pulse frequency and reverse voltage do not appear to influence the response variable, whereas the level average for reverse time varies from 10 000 to virtually zero as this parameter is increased from 1 to 10 μs. Clearly, varying the reverse, or pulse-off time can have a very significant

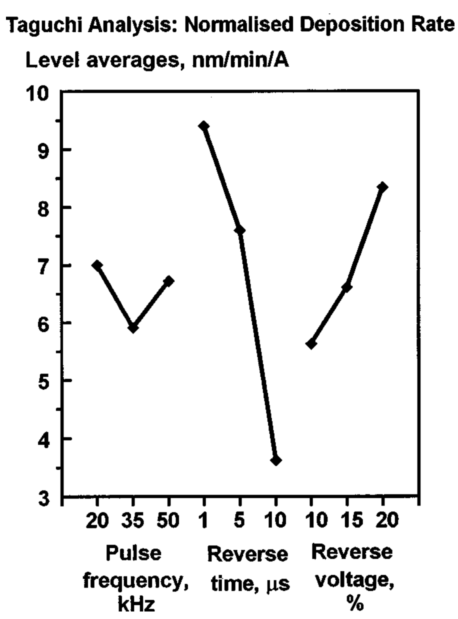

FIG. 5. Taguchi analysis of alumina films, using the normalized deposition

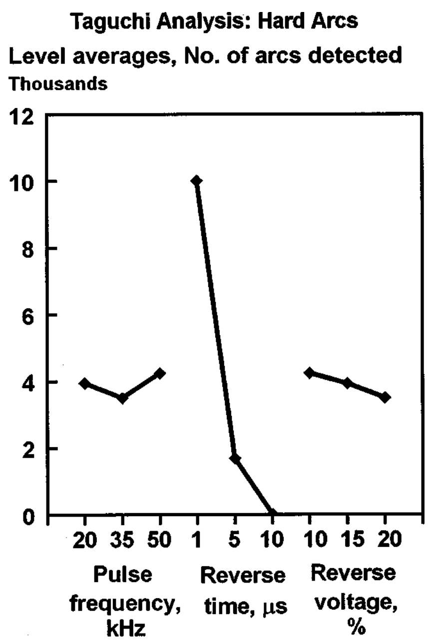

FIG. 7. Taguchi analysis of alumina films, using the total number of hard

DOCKET ALARM

Explore Litigation Insights

Docket Alarm provides insights to develop a more informed litigation strategy and the peace of mind of knowing you're on top of things.

Real-Time Litigation Alerts

Keep your litigation team up-to-date with **real-time alerts** and advanced team management tools built for the enterprise, all while greatly reducing PACER spend.

Our comprehensive service means we can handle Federal, State, and Administrative courts across the country.

Advanced Docket Research

With over 230 million records, Docket Alarm's cloud-native docket research platform finds what other services can't. Coverage includes Federal, State, plus PTAB, TTAB, ITC and NLRB decisions, all in one place.

Identify arguments that have been successful in the past with full text, pinpoint searching. Link to case law cited within any court document via Fastcase.

Analytics At Your Fingertips

Learn what happened the last time a particular judge, opposing counsel or company faced cases similar to yours.

Advanced out-of-the-box PTAB and TTAB analytics are always at your fingertips.

API

Docket Alarm offers a powerful API (application programming interface) to developers that want to integrate case filings into their apps.

LAW FIRMS

Build custom dashboards for your attorneys and clients with live data direct from the court.

Automate many repetitive legal tasks like conflict checks, document management, and marketing.

FINANCIAL INSTITUTIONS

Litigation and bankruptcy checks for companies and debtors.

E-DISCOVERY AND LEGAL VENDORS

Sync your system to PACER to automate legal marketing.

WHAT WILL YOU BUILD? | **sales@docketalarm.com** | **1-866-77-FASTCASE**

fastcase®
Smarter legal research.