Effective Closed-Loop Control for Reactive Sputtering Using Two Reactive Gases

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ABSTRACT

Reactive sputtering is becoming a more widely accepted practice for the deposition of many useful compound thin films. The use of closed-loop control for the deposition of such films is also increasing in popularity. The closed-loop process for forming binary compounds, typically oxides or nitrides, is conveniently accomplished using one of several feedback mechanisms coupled to a high-speed reactive gas control valve. Optical emission, target voltage and mass spectroscopy have all been successfully used to provide feedback for such processes when using a single reactive gas. The presence of two reactive gases in a reactive sputtering process adds significant complexity and presents the issue of competing reactions. Since both reactive gases can affect the state of the target surface and the plasma conditions, both also affect common feedback control signals such as the cathode voltage and optical emission. Modeling has shown that the way to control a two-gas reactive sputtering process is to produce individual control signals for each gas. In this study, partial pressure signals for each reactive gas were used to control the deposition of SiO_xN_y and TiO_xN_y compounds. The results of this study are presented here and shown to support the modeled behavior. The two-gas, closed-loop approach is then demonstrated to provide a stable, high rate solution for the deposition of these highly complex thin film materials.

INTRODUCTION

Oxide and nitride thin films are becoming increasingly popular for their electrical, optical, wear resistant, and other functional properties. Driven by improvements in deposition techniques, these thin film materials are becoming commonplace in many vacuum coating applications. Reactive sputtering continues to emerge as one of the most economical methods for depositing these materials. With properly configured power delivery and process control measures, high rate, arc free deposition of many oxides and nitrides is now possible. As reactive sputtering methods have matured, interest in extending the technique beyond simple binary compounds has been growing. As material complexity increases however, so do the challenges of process control as we examine and discuss in this study.

Although the concept of reactive sputtering is quite simple, the art of employing it effectively to produce high quality insulating films at maximum rates can be a real challenge. The nature of forming insulating layers in a direct-current sputter deposition is inherently disruptive to the process. In recent years the issue of arcing in reactive sputtering has been effectively managed using one of several power delivery techniques [1-4]. The most common of these are pulsed-dc in a single magnetron system and low frequency ac power in a dual magnetron system. In both cases a voltage reversal at the cathode is used to effectively eliminate charge build up on the target surface and thus avoid the breakdown event. Many modern power supplies also possess arc detection and arc handling circuitry designed to quickly extinguish an arc should one occur.

Target transition control is another major challenge in these processes. As the reacted insulating deposit is formed on the work piece, it also forms on the target surface causing a change in its sputtering character. Increasing reactive gas flow to the sputtering process increases conversion of the deposit to the desired compound but also increases the coverage percentage of the compound on the surface of the target. At a critical point, an avalanche transition often occurs where full coverage of the target is experienced. This change in target condition, commonly called poisoning, has several detrimental effects on the process. Dramatic voltage swings, a decrease in deposition rate and an uncontrolled increase in processing pressure are all common results of target poisoning [5-7]. Further complicating the matter is the hysteresis character commonly brought on by this behavior. Figure 1 shows a typical transition curve for reactively sputtered titanium nitride showing how the poisoning process is not directly reversible by simply decreasing reactive gas flow.

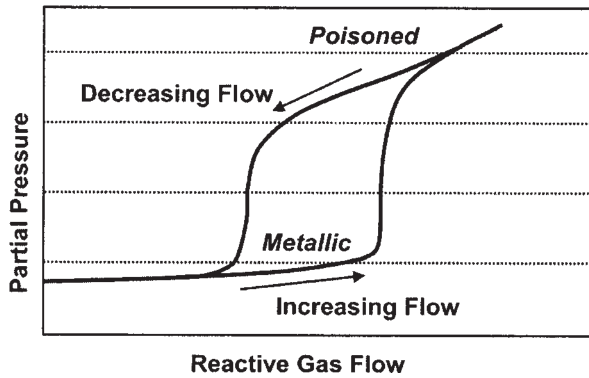

Figure 1: Partial pressure hysteresis behavior typical for titanium nitride reactive sputtering.

Reactive sputtering can be performed from a fully poisoned target but this approach results in low deposition rate and lack of composition control. Operation of these processes in the transition region offers a significant rate advantage [7-9] along with the ability to control the composition of the produced film [9]. This would be most advantageous in a two reactive gas process as the potential exists for a broad range of film properties by adjusting, for instance, the ratio of oxygen and nitrogen incorporated in an oxynitride film. Compositional control in oxynitride depositions has been demonstrated using CVD, PECVD and ion assisted evaporation [10-12] but these techniques tend toward higher complexity and cost and often require more heat tolerant substrates than most typical sputtering applications.

Over the past two decades closed-loop control systems have been developed for reactive sputtering processes [5,6]. These techniques, when properly implemented, allow for stable control within the so-called transition region of the target. Process feedback is in the form of optical emission, mass spectrometry or target voltage [5,6,9] and control is derived by regulation of a single reactive gas through one or more high speed flow devices.

Adding a second reactive gas significantly complicates the process control challenge [13]. With two reactive gases present, the independent reactions compete with one another. Depending on the reactivity between the metal and each gas, one reaction may dominate causing the process to become trapped in an undesirable state preventing incorporation of the less dominant reactive gas. This behavior was predicted in the Carlsson model and has subsequently been demonstrated experimentally [14,15]. To prevent this, independent control of both reactions is necessary.

Control in one gas reactive depositions is often achieved by managing the surface condition of the target. Target voltage is convenient when transition causes an impedance change and optical emission of the sputtered species is effective when transition affects sputter yield. These methods, however, offer no means for discriminating the effects of multiple reactive gases. To accomplish such discrimination the partial pressure of each reactive gas must be measured. As compound formation consumes a reactive gas, the partial pressure is affected. During target transition, the consumption of reactive gas decreases due to reduced unreacted target surface and decreased metal flux to other surfaces. This behavior gives rise to partial pressure monitoring as a direct feedback for compound formation in such a process. Most importantly, partial pressure control offers the critical advantage of being species dependent, thus allowing for independent monitoring and control of multiple reactive species.

Depositions for this study were performed in an open volume, batch style, cylindrical chamber measuring approximately 50 cm in diameter and 55 cm deep. An Angstrom Sciences 150 mm source was used with either a silicon or titanium metallic target. The chamber was pumped to a base pressure of less than 1.33 x 10^{-4} Pa (1.0 x 10^{-6} Torr) prior to each test and operated with 0.4 Pa (3.0 x 10^{-3} Torr) argon. An Advanced Energy Pinnacle®Plus pulsed-dc power supply was operated at 1000 watts pulsed at 100 kHz and 60 or 80% duty factor for all depositions (depending on arc activity). An Advanced Energy IRESS reactive sputtering controller [16] equipped with an Inficon Transpector 2 mass spectrometer was used to actively regulate reactive gas partial pressures by providing control signals to Advanced Energy Aera® 980 series high-speed mass flow controllers. Glass slides and polished silicon slices were used as substrates and loaded onto a shuttered, rotating sample holder. A schematic of the test setup is shown in Figure 2.

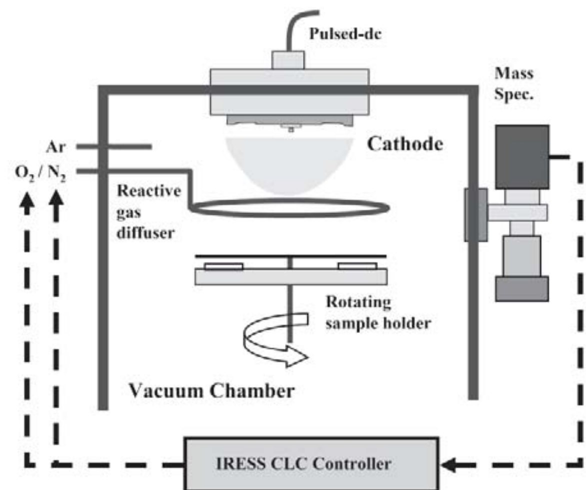

Figure 2: Vacuum and control apparatus used for two gas reactive sputtering process.

Using partial pressure reactive gas control we first investigated single gas behaviors, depositing oxides and nitrides. We evaluated target transition effects with each reactive gas and then showed how adding a second gas using flow control only can lead to unwanted transitions and target trapping. The effect of adding partial pressure control for the second reactive gas was then investigated demonstrating a means for stable control throughout a broad processing space allowing access to a wide operating range and expansive compositional matrix.

RESULTS

Two Gas Reactive Sputtering for Silicon Oxynitride

Oxide and nitride transition curves were generated in order to verify operation and baseline the process in preparation for multi-gas processes. Figure 3 shows reactive gas partial pressure versus reactive gas flow for silicon oxide and silicon nitride formation. Partial pressure values are reported on a 0-10 volt scale representing current collected on the Faraday cup in the mass spectrometer. These values are proportional to the actual pressure of each constituent being measured. Pumping curves for both reactive gases are also shown for reference. Noteworthy differences are the relatively higher pumping efficiency for nitrogen and increased consumption of oxygen to the reaction, especially at the lower partial pressures. This gives rise to a maximum flow value of oxygen at the onset of target transition of approximately 15 sccm. A maximum flow and indication of transition are not observed for the nitrogen reaction.

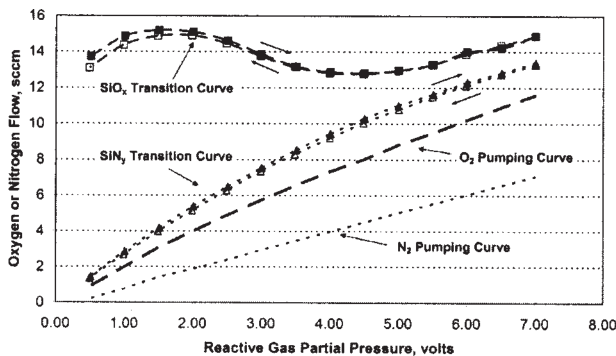

Figure 3: Partial pressure transition curves for SiO_x and SiN_y. Pumping curves for each gas for comparison.

With partial pressure control in place, all points on each curve in Figure 3 were available for stable operation. In addition, stable control was achieved by either starting in the metal mode or by beginning the process in the poisoned or dielectric mode.

As nitrogen is added to the silicon oxide process the effect is to suppress the amplitude of the transition curve, or decrease the amount of oxygen consumed in the process. Figure 4 shows this effect in a family of curves generated with increasing *fixed* nitrogen flow. In these curves, only oxygen is under partial pressure control. The importance of this distinction is discussed below.

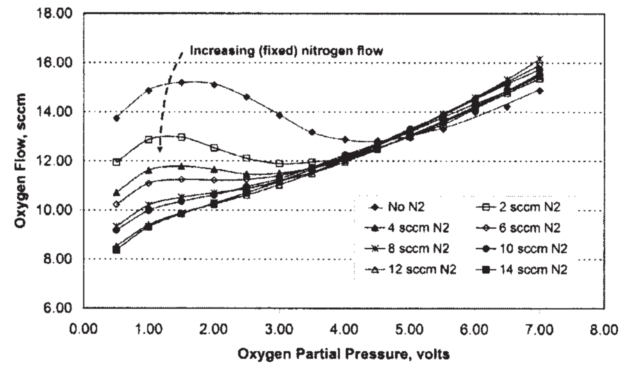

Figure 4: Fixed nitrogen flows and their effect on oxygen partial pressure transition curves for SiO_xN_y.

Above approximately 12 sccm nitrogen, the transition curves in Figure 4 become nearly linear, suggesting that little if any oxygen is being incorporated in the reacted film. Despite the significant change with increased nitrogen flow, stable process control throughout the transition appears to be achieved in each case as the smooth character in the transition is maintained.

The corollary to Figure 4 is to operate the deposition in nitrogen partial pressure control. Here oxygen is added at fixed flow values. Figure 5 gives these results.

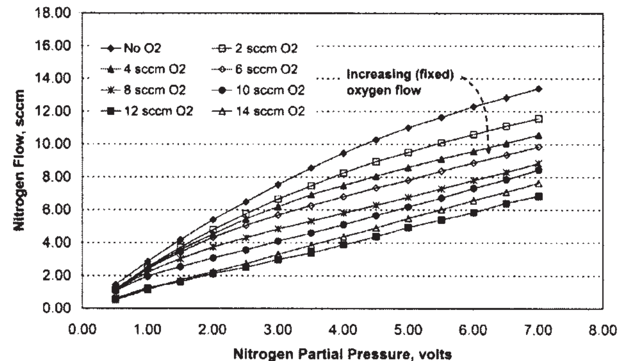

Figure 5: Fixed oxygen flows and their effect on nitrogen partial pressure transition curves for SiO_xN_y.

Again, the curvature is suppressed as the second gas is added indicating reduced nitrogen consumption at higher oxygen flows. Throughout the range tested, the process appears to remain stable. This observation, however, is misleading. What is not shown in Figure 5 is the oxygen partial pressure. For this test only nitrogen was under partial pressure control. In this case oxygen was in fixed flow control leading to the behavior shown in Figure 6.

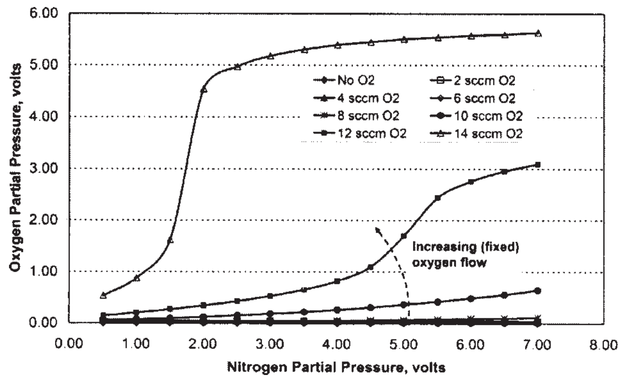

Figure 6: Oxygen partial pressure at fixed flows with nitrogen under partial pressure control.

Figure 6 illustrates the onset of target trapping as described by Carlsson et.al. [13]. At low oxygen flows, consumption by the reaction keeps oxygen partial pressures low throughout the tested nitrogen partial pressure range. As the oxygen flow is increased, the process begins to poison, resulting in an uncontrolled increase in oxygen partial pressure. This behavior indicates the presence of oxygen in excess of what the deposition process can consume.

The trapping effect is more clearly illustrated in Figure 7 where partial pressures of both nitrogen and oxygen were plotted along with target voltage. As before, nitrogen partial pressure is actively controlled and is incrementally increased over time to a predefined maximum. At this point the nitrogen partial pressure is decreased back to zero. As before, oxygen is introduced at a fixed flow and its partial pressure is monitored but not directly controlled. The oxygen flow is 14 sccm here, chosen based on results from Figure 6.

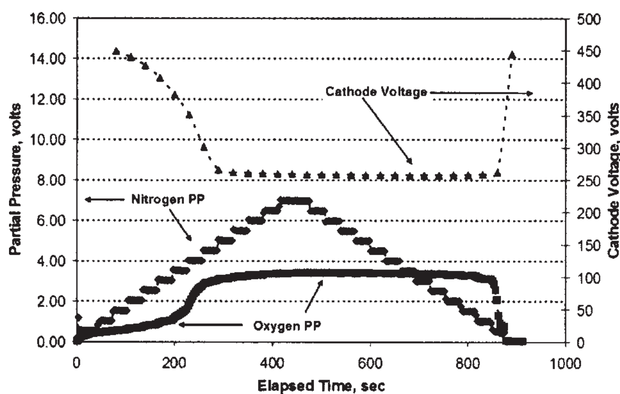

Figure 7: Cathode voltage and reactive gas partial pressures during SiO_xN_y deposition resulting in target trapping. Nitrogen is under partial pressure control and oxygen fixed at 14 sccm.

As nitrogen partial pressure increases we initially observe a gradual increase in oxygen partial pressure. At just over three minutes into the run, a poisoning event is experienced causing a rapid increase in oxygen partial pressure and a rapid decrease in sputtering voltage. Once poisoned, the oxygen partial pressure remains relatively unchanged at a point of saturation and the target remains in a trapped state until both gases are removed from the process.

We found that trapping can be prevented by applying partial pressure control to both reactive gases. Figure 8 gives results from the same process sequence carried out in Figure 7, but this time operated with partial pressure control for oxygen as well. In this case oxygen partial pressure was set to 50% the nitrogen partial pressure.

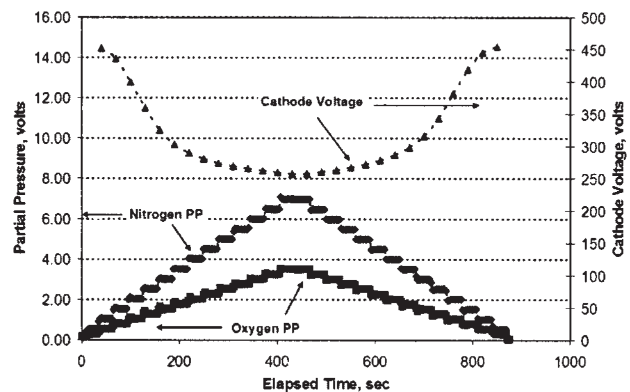

Figure 8: Cathode voltage and reactive gas partial pressures during deposition of SiO_xN_y using partial pressure control of both gases. No target trapping is observed.

Similarly smooth transitions, not shown here, were obtained across a broad range of oxygen to nitrogen partial pressure ratios (O_2:N_2) from 0.05:1 to 4:1, operated with either nitrogen or oxygen as the primary gas. None of these runs, operated with both gases in partial pressure control, showed any evidence of uncontrolled target poisoning or trapping.

With stable control established, we tested the ability to deposit intermediate oxynitride compositions. Figure 9 gives rate and refractive index measurements for films deposited with varying reactive gas partial pressure ratios. At either extreme in the chart we see results representative of the binary process. SiO_x films yielded RI ~ 1.45 at 1510 Å/min while SiN_y gave RI ~ 2.10 at 940 Å/min. Intermediate compositions provided values between the two extremes. These results demonstrate the ability to use such control to access intermediate compositions possessing properties between those of the pure oxide or nitride film.

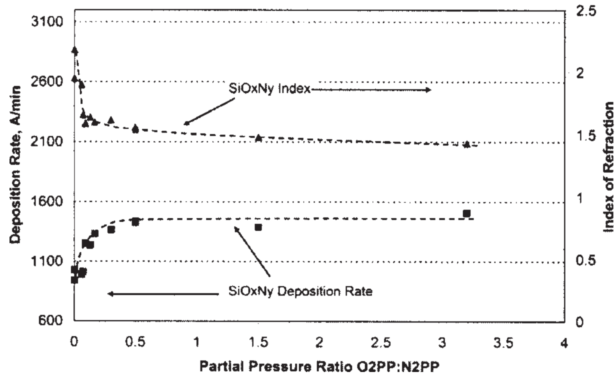

Figure 9: Rate and refractive index for SiO_xN_y deposited by reactive gas partial pressure control.

Two Gas Reactive Sputtering for Titanium Oxynitride

The titanium oxynitride process shows many similarities to the silicon process. The most notable difference is a rise in cathode voltage upon target transition opposed to the fall seen for silicon. Aside from this difference, most other behaviors are quite similar. Titanium oxide and nitride transition curves are shown in Figure 10. As was the case for silicon, the oxide process consumes more reactive gas and shows a more distinct target transition.

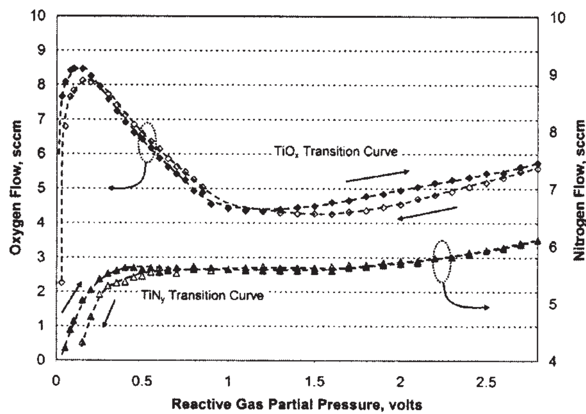

Figure 10: Partial pressure transition curves for TiO_x and TiN_y.

Target trapping was also observed for titanium oxynitride depositions. Again, as we saw for silicon, when one of the gases is regulated by flow only, the possibility for trapping exists. Figure 11 shows a case where we controlled the nitrogen partial pressure and operated oxygen at a fixed flow of 7 sccm. For this test nitrogen partial pressure was increased incrementally and then decreased. A poisoning event is clearly shown at approximately 400 seconds into the run. From this point the target is trapped as indicated by both oxygen partial pressure and target voltage. Only when the reactive gases are removed does the target transition out of the poisoned state.

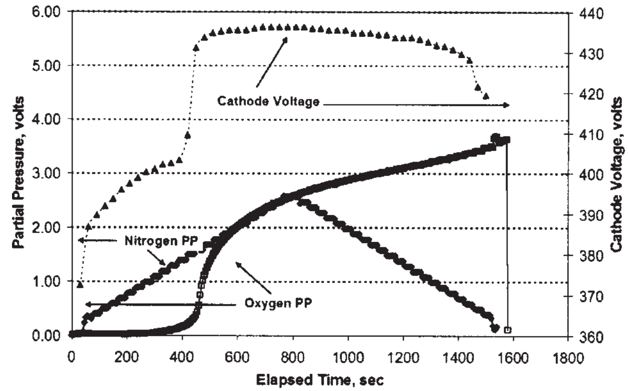

Figure 11: Cathode voltage and reactive gas partial pressures during TiO_xN_y deposition resulting in target trapping. Nitrogen is partial pressure controlled and oxygen is fixed at 7 sccm.

Partial pressure control of both reactive gases was again shown to prevent this behavior. We tested a wide range of partial pressure combinations and found stable, controlled operation in all cases. Figure 12 is one example of such control. Here both oxygen and nitrogen are subjected to partial pressure control. The result was a smooth transition throughout the entire range tested for both reactive gases as well as for target voltage.

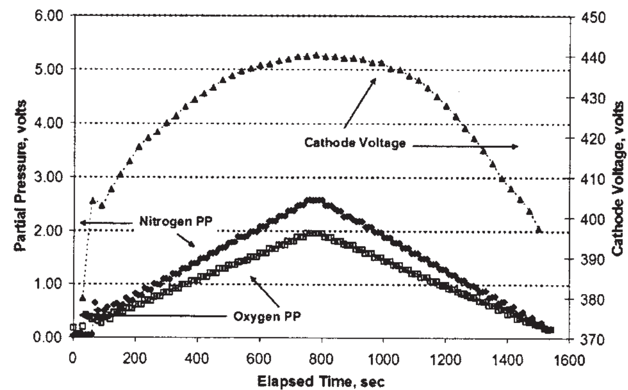

Figure 12: Cathode voltage and reactive gas partial pressures during deposition of TiO_xN_y using partial pressure control of both gases. No target trapping is observed.

DISCUSSION

The presence of multiple reactive gases in an oxynitride reactive sputtering process adds variables that can exceed the control capability of the typical single gas technique. The multi-gas process is complicated as the two reactive gases compete for available metal. The two simultaneous reactions taking place do not act independent of one another and, in fact, one reaction may dominate allowing the process to become poisoned and then trapped in an undesirable state preventing stable operation of the process.

To effectively prevent target trapping, a process control technique capable of differentiating the effects of individual reactive gases is needed. We have demonstrated the occurrence of target trapping in both silicon oxynitride and titanium oxynitride reactive depositions. We also show the behavior to be effectively prevented by employing partial pressure control for both the oxygen and nitrogen reactive gases during deposition. By using such control, a broad range of operating conditions is available providing stable process performance throughout.

Generating a composite summary of the operating range tested, we can produce a process response surface for stable operation of such depositions. One such response surface is shown in Figure 13 for the silicon oxynitride deposition studied.

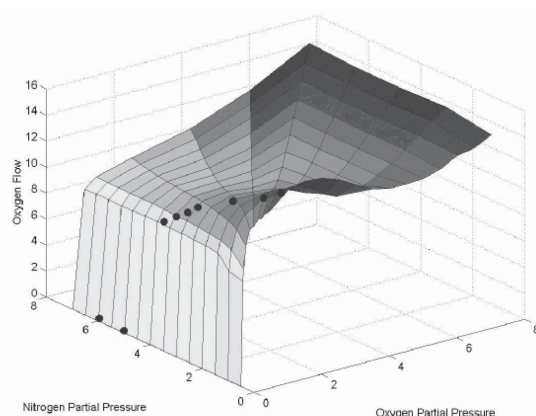

Figure 13: Response surface for reactive deposition of silicon oxynitride using partial pressure reactive gas control.

Using this surface one can visualize a vast range of operating conditions available to perform the two gas reactive deposition. We studied film properties from only a small portion of this surface. Points tested are indicated on the chart and film data were presented earlier, in Figure 9.

CONCLUSION

Measuring the partial pressure of reactive gases provides a critical means for control of individual gases in a two reactive gas deposition. Without the ability to differentiate the effects of each reactive gas such processes are subject to a phenomenon called target trapping. Target trapping can be effectively prevented by controlling the partial pressure of the individual reactive gases in a multi-gas reactive sputtering process. With such control in place, reactive sputtering can be successfully used to produce oxynitride thin films with varying compositions and properties.

REFERENCES

1. S. Schiller, K. Goedicke, J. Reschke, V. Kirchhoff, S. Schneider, and F. Milde, "Pulsed magnetron sputter technology," *Surf. Coat. Technol.*, 61 (1993).

2. P. Kelly, P. Henderson, R. Arnell, G. Roche, D. Carter, "Reactive pulsed magnetron sputtering process for alumina films," *J. Vac. Sci. Technol.*, 18, 6, pp 2890-2896, (2000).

3. G. Este, and W.D. Westwood, "A quasi-direct-current sputtering technique for the deposition of dielectrics at enhanced rates," *J. Vac. Sci. Technol.*, A 6 (3) 1988.

4. D.A. Glocker, "Influence of the plasma on substrate heating during low-frequency reactive sputtering of AIN," *J. Vac. Sci. Technol.* A 11, 2989 (1993).

5. W.D. Sproul, "High rate reactive sputtering process control," *Surf. Coat. Technol.*, 33, 1987.

6. S. Schiller, U. Heisig, Chr. Korndorfer, G. Beister, J. Reschke, K. Steinfelder, and J. Strumpfel, "Reactive d.c. high rate sputtering as a production technology," *Surf. Coat. Technol.*, 33, 1987.

7. S. Berg, H-O. Blom, T. Larsson, and C. Nender, "Modeling of reactive sputtering of compound materials," *J. Vac. Sci. Technol.,* A 5, 1987.

8. J.M. Schneider, W. D. Sproul, M.-S. Wong, and A. Matthews, "Very High Rate Pulsed DC Sputter Deposition of AlOx Coatings," *Surf. Coat. Technol.,* 96 262 (1997).

9. K. Koski, J. Holsa, and P. Juliet, "Voltage controlled reactive sputtering process for aluminum oxide thin films," *Thin Solid Films*, 326, 1998.

10. Y. Ma and G. Lucovsky, "Deposition of single phase, homogeneous silicon oxynitride by remote plasma-enhanced chemical vapor deposition, and electrical evaluation in metal-insulator-semiconductor devices," *J. Vac. Sci. Technol.*, B 12(4), 1994.

11. G.A. Al-Jumaily, T.A. Mooney, W.A. Spurgeon, and M. Dauplaise, "Ion assisted deposition of oxynitrides of aluminum and silicon," *J. Vac. Sci. Technol.*, A 7(3), May/June 1989.

12. J. Vuillod, "Preparation and characterization of plasma enhanced chemical vapor deposited silicon nitride and oxynitride films," *J. Vac. Sci. Technol.*, A 5(4), (1987).

13. P. Carlsson, C. Nender, H. Barankova, and S. Berg, “Reactive sputtering using two reactive gases, experiments and computer modeling,” *J. Vac. Sci. Technol.*, A 11(4), (1993).

14. H. Barankova, S. Berg, P. Carlsson, and C. Nender, “Hysteresis effect in the sputtering process using two reactive gases,” *Thin Solid Films*, 260, p. 181, 1995.

15. W.D. Sproul, D.J. Christie, D.C. Carter, S. Berg, and T. Nyberg, “Control of the Reactive Sputtering Process Using Two Reactive Gases,” *47th Annual Technical Conference Proceedings of the Society of Vacuum Coaters*, 2003.

16. W. D. Sproul and B. E. Sylvia, “An Integrated Solution for Reactive Sputtering,” *Vac. Tech. and Coating*, 2 No. 8, 32, 2001.