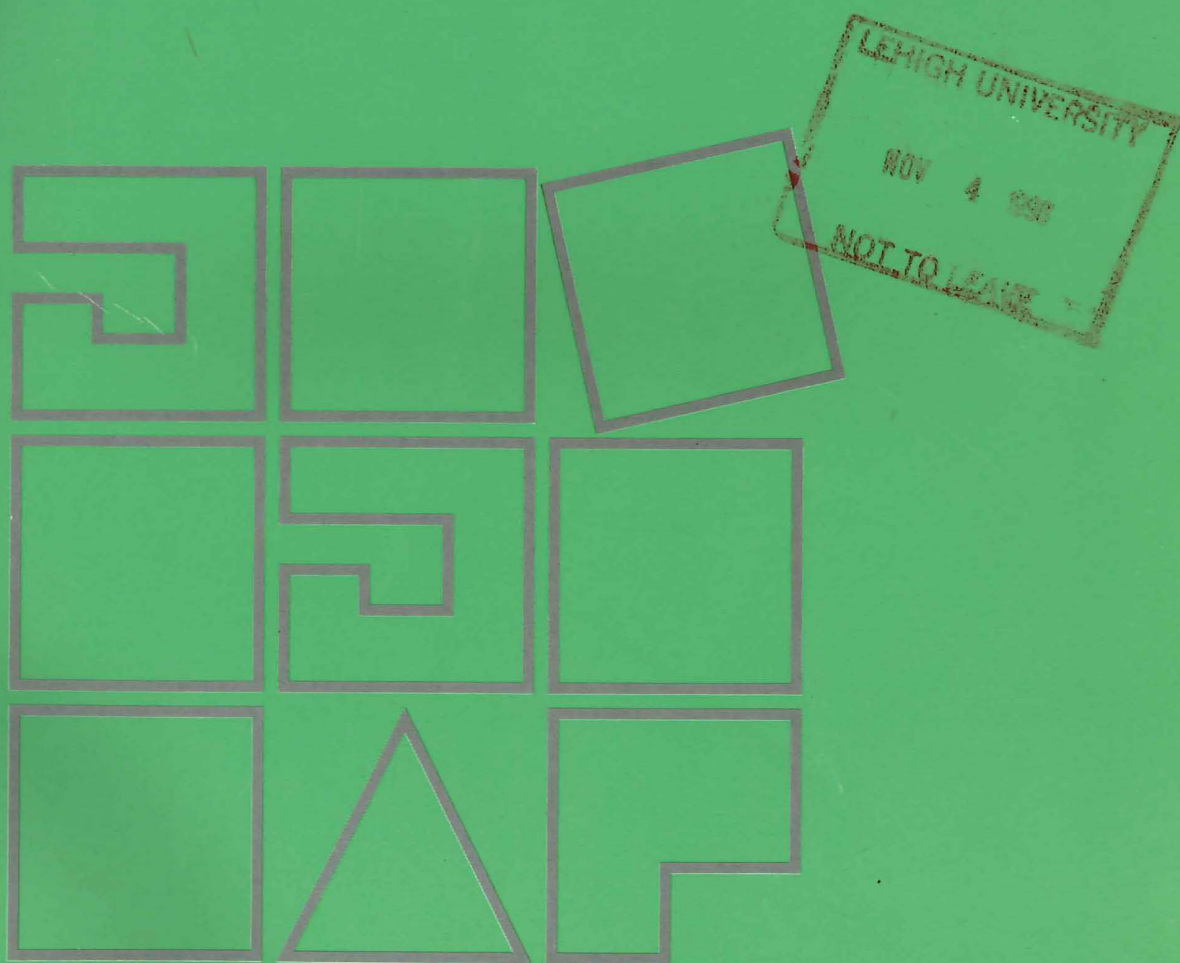


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Atomic Layer Deposition of TiN Films by Alternate Supply of Tetrakis(ethylmethlamino)-Titanium and Ammonia

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Atomic layer deposition (ALD) of amorphous TiN films on SiO₂ between 170°C and 210°C has been investigated by alternate supply of reactant sources, Ti[N(C₂H₅CH₃)₂]₄ [tetrakis(ethylmethlamino)titanium:TEMAT] and NH₃. Reactant sources were injected into the reactor in the following order: TEMAT vapor pulse, Ar gas pulse, NH₃ gas pulse and Ar gas pulse. Film thickness per cycle was saturated at around 1.6 monolayers (ML) per cycle with sufficient pulse times of reactant sources at 200°C. The results suggest that film thickness per cycle could exceed 1 ML/cycle in ALD, and are explained by the chemisorption mechanism of the reactant sources. An ideal linear relationship between number of cycles and film thickness is confirmed. As a result of surface limited reactions of ALD, step coverage was excellent. Particles caused by the gas phase reactions between TEMAT and NH₃ were almost absent because TEMAT was segregated from NH₃ by the Ar pulse. In spite of relatively low substrate temperature, carbon impurity was incorporated below 4 at. %.

KEYWORDS: TiN, atomic layer deposition (ALD), alternate supply, TEMAT

1. Introduction

Conformality of step coverage is an important factor affecting deposition of TiN diffusion barriers during multilevel metal interconnection processes in ultra-large scale integration (ULSI) technology. Generating conformal step coverage has become more difficult as device dimensions have been reduced, increased contact and via hole aspect ratio. TiN chemical vapor deposition (CVD) is one of the most promising methods because it provides much better step coverage for films than conventional TiN sputtering. As a result, considerable research has focused on TiN CVD during the last several years.^{1–3)}

Two types of titanium sources, TiCl₄ and metalorganic precursors have been widely used with NH₃ as reactive gas for TiN CVD. In TiCl₄-based TiN CVD, comparatively high conductive TiN films with good step coverage could be deposited.^{4,5)} However, TiCl₄-based TiN CVD suffers in that the deposition temperatures are too high for the process to be applicable at all metallization levels and serious corrosion of Al occurs, caused by Cl impurities and the formation of adducts and particles such as NH₄Cl salts. Recently, high-quality TiN films resulting from the reaction of metalorganic precursors and NH₃ have been deposited.^{6–8)} It has been understood that TiN films are deposited by the transamination reaction of tetrakis(dialkylamino)titanium [Ti(NR₂)₄] (where R=methyl, ethyl complexes) and NH₃. This process, however, reveals poor step coverage, probably due to the highly reactive intermediates produced in the gas phase. In addition, this process also suffers from particle contamination caused by the gas phase reactions between NH₃ and metal-organic precursors such as tetrakis(dialkylamino)titanium complexes. In order to address these problems encountered with these binary chemistries, recent work on CVD of TiN films has focused on a single-source process using metalorganic precursors.^{8,9)} This process provided good step coverage, however, it produced high resistivity and a high level of carbon contamination. Therefore, a new process needs to be developed to produce high-quality TiN films with improved conformality.

In the present paper, in accordance with the observation of poor step coverage and particle generation caused by gas phase reactions between metal-organic precursors and NH₃ in MOCVD, TiN films have been deposited on SiO₂ by an alternate supply of TEMAT and NH₃ to eliminate their gas phase reactions. Using this approach, it is possible to grow TiN films by the atomic layer deposition (ALD) process.^{10–12)} As a result of the promotion of surface limited reactions of ALD, conformal films can be obtained with less contact and via hole size. Because TEMAT was separated from NH₃ by an Ar pulse, particles usually produced by the gas phase reactions between TEMAT and NH₃ were rarely detected. In spite of relatively low substrate temperature ($\leq 200^\circ\text{C}$), a low level of carbon was incorporated by transamination reactions between TEMAT and NH₃ on the surface. Therefore, the objective of the present work is to produce high-quality TiN films with improved conformality by the ALD process which is crucial for the interconnection of metals in ultralarge-scale integrated circuits.

2. Experimental

TiN films were deposited on SiO₂ by alternate supply of TEMAT and NH₃. Figure 1 schematically illustrates the deposition apparatus which consists of a warm-wall reactor, a gas switching system and a pumping system. Deposition of all TiN films was carried out at substrate temperatures of between 150°C and 400°C and at a pressure of 0.5 Torr. SiO₂(100 nm)/Si(100)-oriented wafers were used. Reactant sources were 250 sccm NH₃ and TEMAT kept at 65°C to maintain a vapor pressure of about 10⁻² Torr, through which 100 sccm H₂ carrier gas was bubbled. 300 sccm Ar gas served as the purge gas.

The reactor and gas lines were sufficiently purged by Ar, assuring that residual oxygen and hydrocarbon molecules were minimized. Reactant sources were injected into the reactor in the following order: TEMAT vapor pulse, Ar purge gas pulse, NH₃ gas pulse and Ar purge gas pulse. These four pulses were defined as one cycle. This cycle was repeated until the thickness was obtained. Figure 2 shows the gas switching method which sequentially injects the reactant

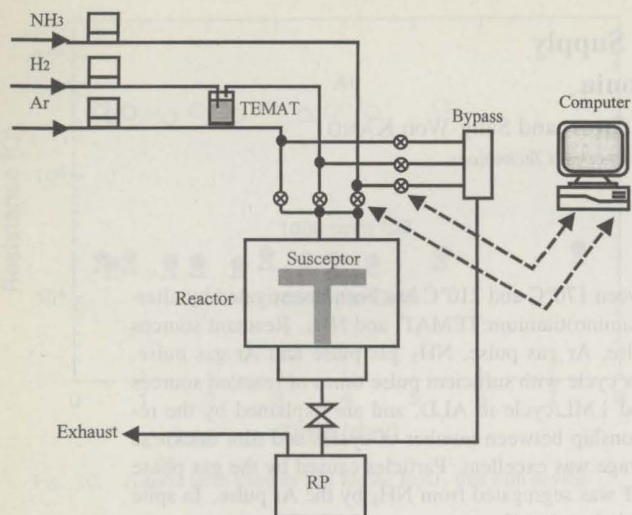
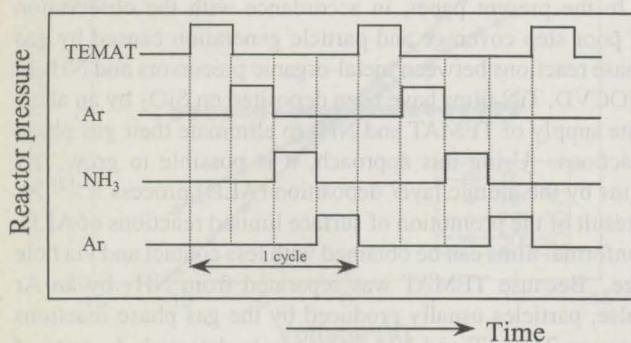


Fig. 1. A schematic diagram of the TiN deposition apparatus.

Fig. 2. Gas switching method which sequentially injects reactant sources and Ar gas with pulse time. TEMAT is introduced with 100 sccm H₂ carrier gas, purged with 300 sccm Ar gas, thus 250 sccm NH₃ is introduced and purged with 300 sccm Ar gas. This periodic cycle is repeated until the desired film thickness is obtained.

sources and Ar for each pulse. In order to clarify the TiN growth mechanism, pulse times of reactant sources were varied between 1 s and 40 s. In this work, a standard cycle was TEMAT vapor pulse of 5 s, post-TEMAT Ar purge of 5 s, NH₃ gas pulse of 10 s and post-NH₃ Ar purge of 5 s, represented simply by (5'', 5'', 10'', 5'').

The sheet resistance and film thickness were measured by the four-point probe and the surface profilometer, respectively. Growth rate was defined in terms of film thickness per cycle and calculated by dividing the total TiN film thickness deposited by the total number of cycles (150 cycles). Scanning electron microscopy (SEM) was used to determine the step coverage. Impurity incorporation and surface density were analyzed by elastic recoil detection-time of flight (ERD-TOF) and Rutherford backscattering spectroscopy (RBS). An X-ray diffractometer (XRD) were employed to determine the crystal structure.

3. Results and Discussion

A definite temperature range for TiN ALD is required not only because the TEMAT vapors are not allowed to decompose thermally, but also because the chemisorbed monolayer

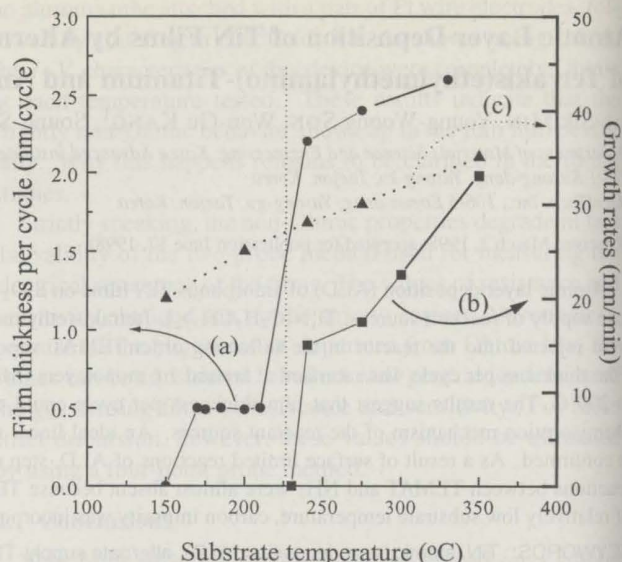


Fig. 3. The dependence of growth rates on substrate temperatures at 0.5 Torr.

- (a) The approach by alternate supply of TEMAT and NH₃, one cycle consists of (15'', 5'', 10'', 5'')
- (b) Conventional MOCVD using TEMAT only without NH₃
- (c) Conventional MOCVD using TEMAT with NH₃

is formed on the surfaces. Also, TiN must be formed by the TEMAT-chemisorbed chemical reactions with NH₃ on the surfaces. It is important to find out the optimal temperature range for TiN ALD, thus, the dependence of growth rates on substrate temperatures is shown in Fig. 3. In (a), TiN films were deposited by an alternate supply of TEMAT and NH₃, where the growth rates are indicated by the film thickness per cycle at the left axis. Cases (b) and (c) were carried out by conventional MOCVD which continuously injects the reactant sources into the reactor during the deposition. TEMAT (b) and TEMAT and NH₃ (c) were used as reactant sources. The temperature range for TiN ALD is listed in Fig. 3. The results of (a) reveal two distinct regions; the first occurs where film thickness per cycle is saturated around 0.5 nm/cycle between 170°C and 210°C, and the other tends to gradually increase above 240°C. By combining results from (a) and (b), it is shown that the transition temperature from ALD to MOCVD is around 230°C. Therefore, the temperature range for TiN ALD is limited to below 230°C. Although TiN films were deposited using TEMAT and NH₃ in both (a) and (c), different deposition results are observed. From the result of (c), growth rates increase as a function of substrate temperature between 150°C and 350°C. However, in case of (a), film thickness per cycle is independent of substrate temperature between 170°C and 210°C. The result of (c) is due to gas phase reactions where the addition of NH₃ to TEMAT leads to facile transamination reactions. In (a), TiN films are not deposited below 150°C, indicating that reactant sources are mainly physisorbed on surfaces below 150°C. Chemisorption involves the formation of relatively strong chemical bonds, whereas in physisorption, the effective forces are of the weak van der Waals type.¹²⁾ Thus, weak bonds of physisorption can be easily purged by Ar. It can be concluded that the temperature range for TiN ALD is between 170°C and 210°C.

To determine the optimal parameters for self-controlled

growth by adsorption of TEMAT, the TEMAT pulse time was varied from 1s to 15s. Figure 4 illustrates the dependence of film thickness per cycle on the TEMAT pulse time at 175°C, 200°C and 240°C. At the substrate temperature of 240°C, film thickness per cycle increases with increasing TEMAT pulse time, which is quite different from the growth at 175°C and 200°C. This is obviously due to self-thermal decomposition of TEMAT on the surfaces. At 175°C and 200°C, which are lower than TEMAT self-decomposition temperature of 230°C, film thickness per cycle is saturated at around 0.5 nm/cycle. After saturation level is reached, film thickness per cycle remains constant, and saturation level is independent of both the TEMAT pulse time and the substrate temperature. These results suggest realization of a self-limiting process by adsorption of TEMAT and are distinct characteristic of ALD process. Also, film thickness per cycle is saturated when the TEMAT pulse time exceeds 3s at 200°C and 10s at 175°C, respectively. The difference in TEMAT pulse length required to reach saturation level between 175°C and 200°C is probably due to temperature dependence on adsorption rate of the reactant sources. Adsorption rate is described by following eq. (1) at constant pressure.¹³⁾

$$\frac{dS}{dt} = A \exp\left(-\frac{E_A}{kT}\right) \quad (1)$$

In this equation, S is coverage, E_A is energy barrier to chemisorption, A is a constant, k is the Boltzmann constant and T is substrate temperature. Because saturated film thickness per cycle is equal to 0.5 nm/cycle at both 175°C and 200°C, it can be assumed that adsorption sites are independent of the substrate temperature. Therefore, adsorption rate increases with an increase in substrate temperature as can be seen from eq. (1). Film thickness per cycle is saturated faster at 200°C than at 175°C.

The relationship between Ar pulse time (1–20s), and activity of gas phase reactions between TEMAT and NH_3 was investigated. Simultaneously, desorption of TEMAT and NH_3

chemisorbed on surfaces by Ar purge was studied. TEMAT pulse time was fixed at 5s because the film thickness per cycle was saturated when the TEMAT pulse time exceeded 3s at 200°C. Figure 5 shows the dependence of film thickness per cycle on the post-TEMAT Ar purge time at 200°C. Film thickness per cycle was saturated at around 0.5 nm/cycle when post-TEMAT Ar purge time exceeded 3s. This suggests that the gas phase reactions between TEMAT and NH_3 can be inhibited and TEMAT chemisorbed on surfaces is not desorbed by Ar purging. Figure 6 shows the dependence of film thickness per cycle on the post- NH_3 Ar purge time at 200°C. Film thickness per cycle is saturated at around 0.5 nm/cycle

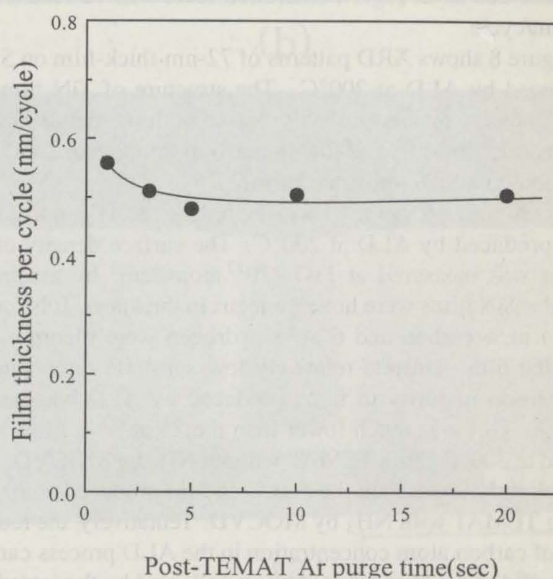


Fig. 5. Film thickness per cycle as a function of Ar pulse time for film grown on SiO_2 at 200°C, 0.5 Torr. One cycle consists of (5'', 1''–20'', 10'', 5'').

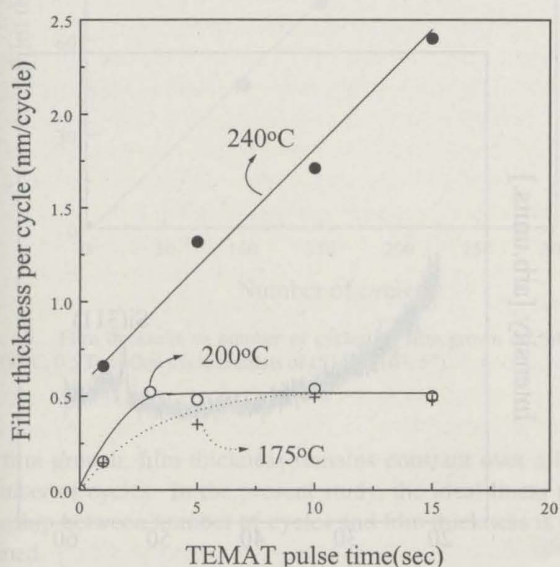


Fig. 4. Film thickness per cycle as a function of TEMAT pulse time for film grown on SiO_2 at substrate temperature 175°C, 200°C and 240°C and pressure of 0.5 Torr. One cycle consists of (1''–15'', 5'', 10'', 5'').

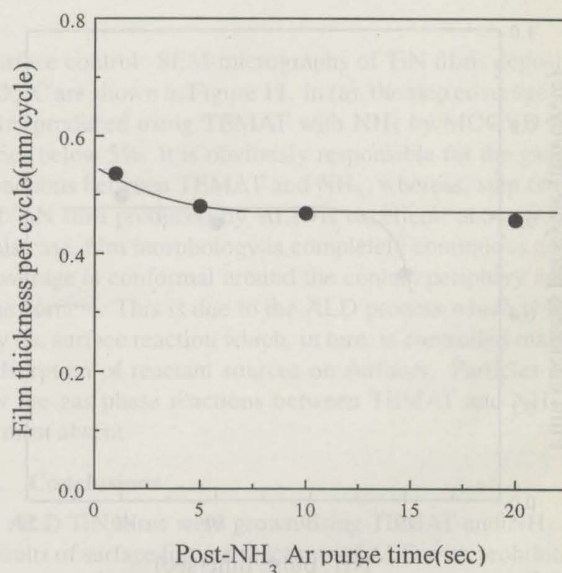


Fig. 6. Film thickness per cycle as a function of Ar pulse time for film grown on SiO_2 at 200°C, 0.5 Torr. One cycle consists of (5'', 5'', 10'', 1''–20'').

i.e., the same results as these of Fig. 5, indicating that the NH_3 chemisorbed on surfaces is not desorbed by the Ar purge. Therefore, when Ar purge time is greater than 5s, the effects of the gas phase reactions and desorptions of adsorbates on film growth can be neglected.

To find the optimal parameters for self-controlled growth by adsorption of NH_3 , the NH_3 pulse time was varied from 10s to 40s. Figure 7 illustrates the dependence of film thickness per cycle on NH_3 pulse time at 200°C . Film thickness is 0.5 nm/cycle when NH_3 pulse time is 10s. At NH_3 pulse times greater than 10s, it is increased slightly with increasing NH_3 pulse time and saturated at around 0.6 nm/cycle when NH_3 pulse time exceeds 20s. This is probably due to the insufficient NH_3 pulse time. In principle, if NH_3 pulse time exceeds 20s as in Fig. 4, saturation level will be reached at 0.6 nm/cycle.

Figure 8 shows XRD patterns of 72-nm-thick-film on SiO_2 produced by ALD at 200°C . The structure of TiN films is amorphous, which is probably due to both the deficiency of thermal driving force at low deposition temperature and deposition on a SiO_2 substrate surface.

Figure 9 illustrates the ERD-TOF spectra of 72-nm-thick-film produced by ALD at 200°C . The surface density of Ti atoms was measured at 1.47×10^{17} atoms/ cm^2 by assuming that the TiN films were homogeneous in thickness. It is found that 4 at.% carbon and 6 at.% hydrogen were incorporated into the film. Despite relatively low substrate temperature, the carbon impurity in films produced by ALD was below 4 at.%. This was much lower than the 25 at.% in films produced at 300°C using TEMAT without NH_3 by MOCVD, and was slightly lower than the 5 at.% in film produced at 200°C using TEMAT with NH_3 by MOCVD. Tentatively, the reduction of carbon atom concentration in the ALD process can be attributed to the post-purge process followed by the introduction of reactant gas.

It is found that saturated film thickness is 0.6 nm/cycle with sufficient pulse times of reactant sources as illustrated

in Fig. 7. To investigate whether a film thickness of 0.6 nm is equal to 1 ML in TiN films, calculation of the distance between subsequent Ti layers is required. The XRD results in Fig. 8 confirm that the structure of the film is amorphous, therefore, it can be assumed that TiN films are isotropic. If we assume that the unit volume of TiN film is cubic, the average distance between Ti atoms can be represented as

$$d_{\text{Ti}} = \left(\frac{t}{\rho_{\text{s,Ti}}} \right)^{\frac{1}{3}} \quad (2)$$

where $\rho_{\text{s,Ti}}$, surface density of Ti atoms, is 1.47×10^{17} atoms/ cm^2 , which was measured by RBS and ERD-TOF in Fig. 9. Film thickness, t , is 72 nm, which was measured by using a surface profilometer. The result of computation of the average distance between Ti atoms d_{Ti} is 0.36 nm. This distance can be considered as a film thickness of 1 ML. As shown in Fig. 7, saturated film thickness per cycle is 0.6 nm/cycle, which corresponds to 1.6 ML/cycle. However it has previously been reported that film thickness per cycle of the ALD process is limited to below 1 ML/cycle.¹⁴⁻¹⁶ It seems that this inconsistency is due to film thickness per cycle in TiN ALD depending not only on TEMAT pulse time, but also on NH_3 pulse time. When NH_3 is introduced into the reactor, some NH_3 reacts with the TEMAT chemisorbed on surfaces resulting in the formation of TiN films and part of the remaining NH_3 can be rechemisorbed on surfaces. When the reverse case is taken into consideration, the same effects can be satisfied. Rechemisorption of these excess reactant sources contribute to growth in excess of 1 ML/cycle. In principle, film thickness per cycle may be in excess of 1 ML/cycle if all of the reactant sources can be chemisorbed on surfaces, although coverage (θ) of reactant sources is lower than 1 ML, ($\theta \leq 1$). This mechanism is in good agreement with the GaAs growth mechanism reported by Sakuma *et al.*¹⁷

One of the attractive features of the ALD process is that film thickness is controlled simply by number of cycles. Figure 10 shows the dependence of film thickness on the number of cycles at 200°C . As it is expected for the ALD process

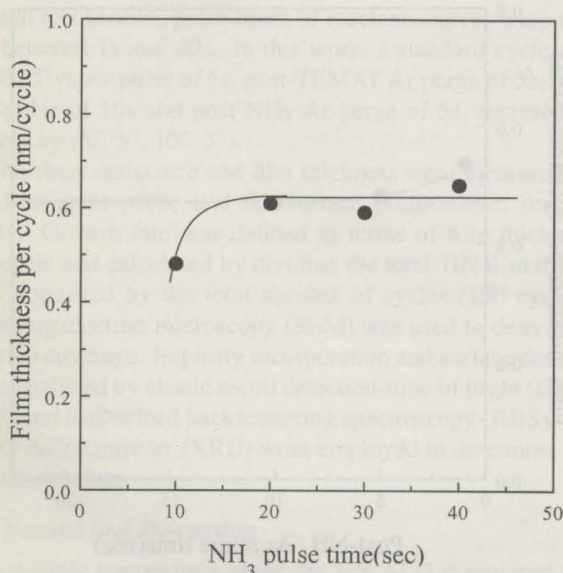


Fig. 7. Film thickness per cycle as a function of NH_3 pulse time for film grown on SiO_2 at 200°C , 0.5 Torr. One cycle consists of (5'', 5'', 10'', 40'', 5'').

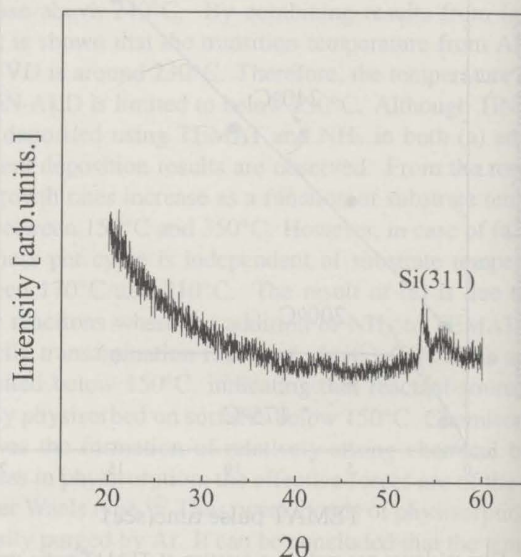


Fig. 8. XRD patterns for film grown on SiO_2 at 200°C , 0.5 Torr. One cycle consists of (5'', 5'', 10'', 5'').

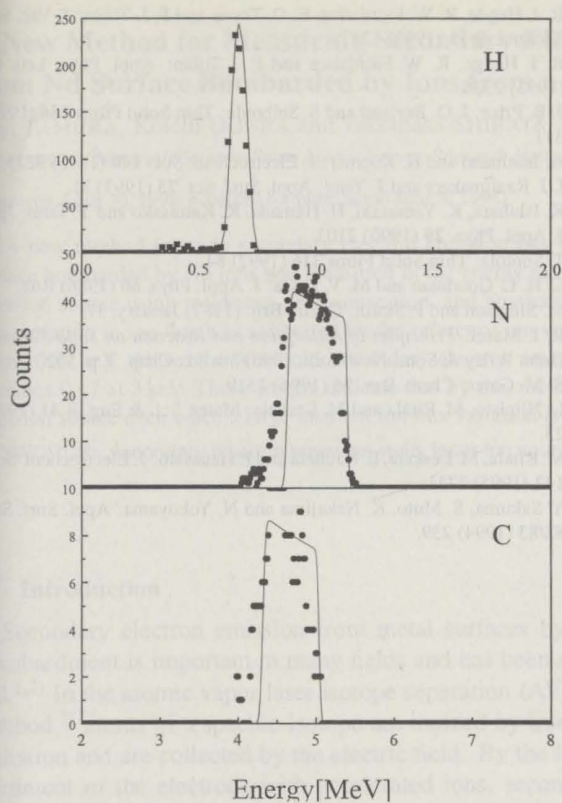


Fig. 9. ERD-TOF spectra of detected elements for film grown on SiO_2 at 200°C , 0.5 Torr. One cycle consists of ($5''$, $5''$, $10''$, $5''$). ($\text{N/Ti} = 0.55$, $\text{C} = 4 \text{ at.}\%$, $\text{H} = 6 \text{ at.}\%$)

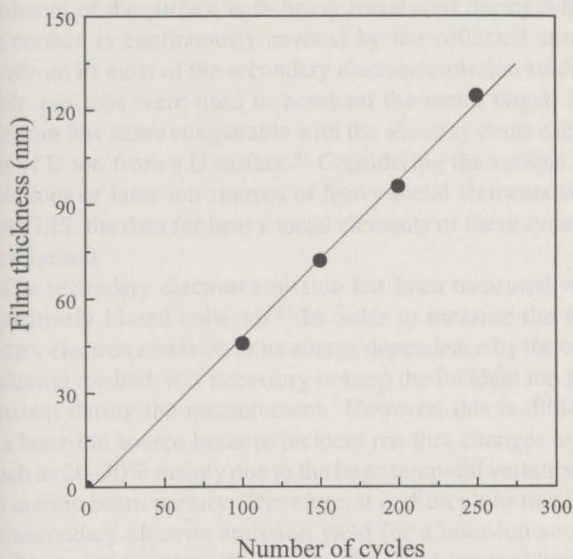


Fig. 10. Film thickness vs number of cycles for film grown on SiO_2 at 200°C , 0.5 Torr. One cycle consists of ($5''$, $5''$, $10''$, $5''$).

of film growth, film thickness remains constant over a large number of cycles. In the present study, the ideal linear relationship between number of cycles and film thickness is confirmed.

The ALD process has great potential for step coverage and conformal coating procedures which are often required in ULSI device manufacturing due to the nature of its required

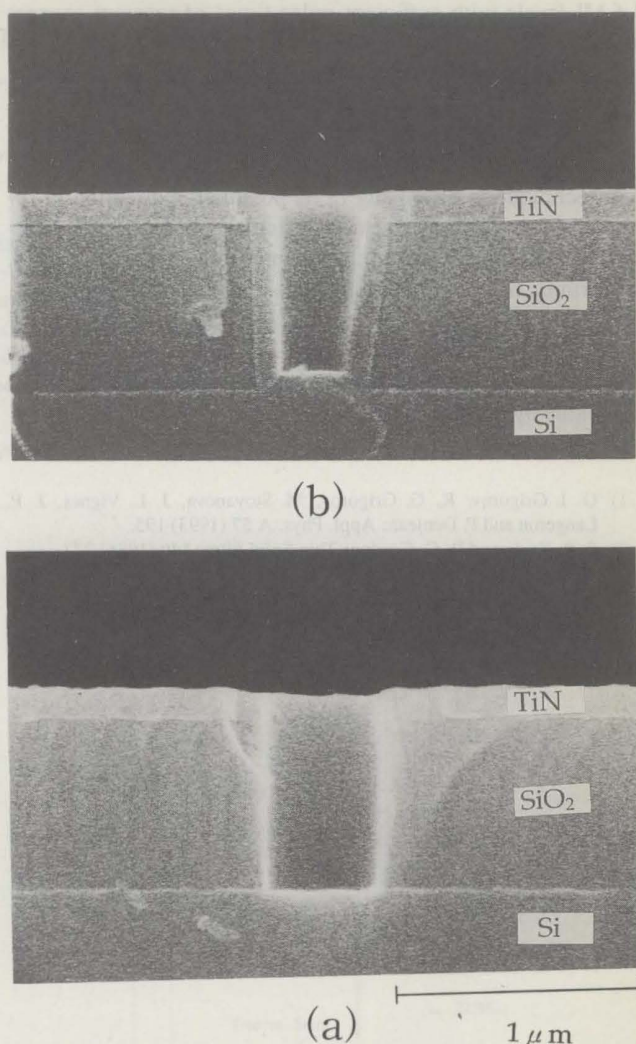


Fig. 11. Cross-sectional SEM micrographs for film grown on SiO_2 at 200°C , 0.5 Torr into $0.6\text{-}\mu\text{m}$ -wide and $0.7\text{-}\mu\text{m}$ -deep contact holes.

(a) Conventional MOCVD using TEMAT with NH_3
(b) ALD; One cycle consists of ($5''$, $5''$, $10''$, $5''$)

surface control. SEM micrographs of TiN films deposited at 200°C are shown in Figure 11. In (a), the step coverage of TiN film produced using TEMAT with NH_3 by MOCVD is very poor below 5%. It is obviously responsible for the gas phase reactions between TEMAT and NH_3 , whereas, step coverage of TiN film produced by ALD is excellent, above 90%. In this case, film morphology is completely continuous and film coverage is conformal around the contact periphery and into the corners. This is due to the ALD process which is limited by the surface reaction which, in turn, is controlled mainly by adsorption of reactant sources on surfaces. Particles caused by the gas phase reactions between TEMAT and NH_3 were almost absent.

4. Conclusions

ALD TiN films were grown using TEMAT and NH_3 . As a results of surface limited reactions of ALD with prohibition of the gas phase reactions between TEMAT and NH_3 , excellent results are useful in the manufacture of ULSI devices.

Film thickness per cycle was saturated at around

1.6 ML/cycle with sufficient pulse times of reactant sources at 200°C. The results, which were explained by rechemisorption mechanisms of reactant sources suggest that film thickness per cycle could be beyond 1 ML/cycle in ALD.

The ALD technique has mainly been utilized for growth of single crystals such as GaAs, however, this new film deposition technique is promising in its potential application in future ULSI device manufacture particularly in terms of conformality, particle elimination, accurate film thickness control, uniformity, reproducibility and low-temperature processing.

Acknowledgments

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- 1) G. I. Grigorov, K. G. Grigorov, M. Stoyanova, J. L. Vignes, J. P. Langeron and P. Denjean: *Appl. Phys. A* **57** (1993) 195.
- 2) S. R. Kurtz and R. G. Gordon: *Thin Solid Films* **140** (1986) 277.
- 3) A. Sherman: *J. Electrochem. Soc.* **137** (1990) 1892.

- 4) R. I. Hegde, R. W. Fiordalice, E. O. Travis and P. J. Tobin: *J. Vac. Sci. Technol. B* **11** (1993) 1287.
- 5) R. I. Hegde, R. W. Fiordalice and P. J. Tobin: *Appl. Phys. Lett.* **62** (1993) 2326.
- 6) J. B. Price, J. O. Borland and S. Selbrede: *Thin Solid Films* **236** (1993) 311.
- 7) A. Intemann and H. Koerner: *J. Electrochem. Soc.* **140** (1993) 3215.
- 8) I. J. Raaijmakers and J. Yang: *Appl. Surf. Sci.* **73** (1993) 31.
- 9) K. Ishihara, K. Yamazaki, H. Hamada, K. Kamisako and Y. Tarui: *Jpn. J. Appl. Phys.* **29** (1990) 2103.
- 10) T. Suntola: *Thin Solid Films* **216** (1992) 84.
- 11) C. H. L. Goodman and M. V. Pessa: *J. Appl. Phys.* **60** (1986) R65.
- 12) M. Simpson and P. Smith: *Chem. Brit.* (1987) January, 37.
- 13) R. I. Masel: *Principles of Adsorption and Reaction on Solid Surfaces* (John Wiley & Sons, New York, 1996) 3th ed., Chap. 7, p. 502.
- 14) S. M. Gates: *Chem. Rev.* **96** (1996) 1519.
- 15) L. Niinisto, M. Ritala and M. Leskela: *Mater. Sci. & Eng. B* **41** (1996) 23.
- 16) M. Ritala, M. Leskela, E. Rauhaluoma and P. Haussalo: *J. Electrochem. Soc.* **142** (1995) 2731.
- 17) Y. Sakuma, S. Muto, K. Nakajima and N. Yokoyama: *Appl. Surf. Sci.* **82/83** (1994) 239.

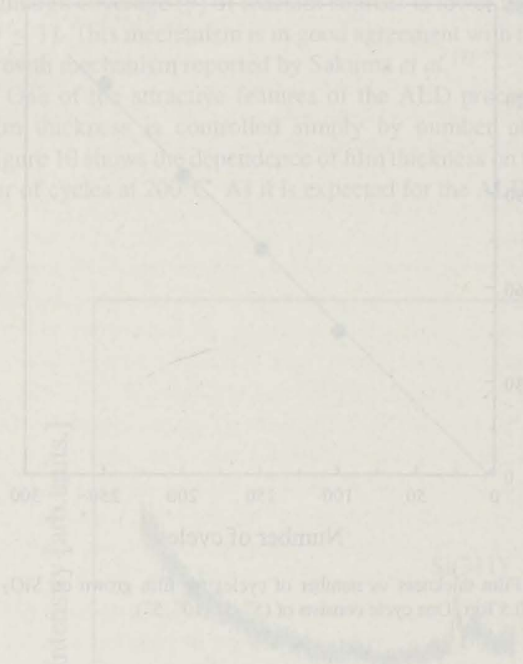


Fig. 1. Relationship between the number of cycles and the film thickness. The film thickness was measured by XPS. The growth rate of the film was about 17 nm/cycle. The error bar is about 10 nm.