Bottom-up plasma-enhanced atomic layer deposition of SiO₂ by utilizing growth inhibition using NH₃ plasma pre-treatment for seamless gap-fill process

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The design-rule shrinkage in semiconductor devices is a challenge at every step of the integration process. In the gap-fill process for isolation, the seam and void formation cannot be suppressed by using a deposition process, which even has excellent step coverage. To achieve seamless gap fill in the high-aspect-ratio structure, which has a non-ideal etch profile such as a negative slope, the deposition process should be able to realize the “bottom-up growth” behavior. In this work, the bottom-up growth of a SiO₂ plasma-enhanced atomic layer deposition (PE-ALD) process in a trench structure was investigated by using a growth inhibition process employing plasma treatment. N₂ and NH₃ plasma pre-treatments were employed to suppress the growth of the SiO₂ PE-ALD process without any contamination, and the inhibition mechanism was investigated by performing surface chemistry analyses using X-ray photoelectron spectroscopy. Furthermore, the gap-fill characteristics of the SiO₂ PE-ALD process were examined, depending on the process conditions of NH₃ plasma pre-treatment, by performing cross-sectional field emission scanning electron microscopy measurements. Finally, a seamless gap-fill process in a high-aspect-ratio trench pattern was achieved by the bottom-up growth behavior of SiO₂ PE-ALD using NH₃ plasma pre-treatment.

Design rule shrinkage is a crucial requirement for increasing the integration density in semiconductors. To achieve a device with a reduced design rule, not only the patterning processes, such as lithography and etching, but also the isolation between actives, where the channel is located, is important to ensure robust device operation. In this regard, isolation processes, such as shallow trench isolation, contact holes, channel hole oxides, and inter-metal dielectrics, have become important for obtaining cutting-edge semiconductor devices. However, the difficulty of the isolation process increases with an increased aspect ratio of the structure and decreased space between actives (less than tens of nanometers) having a fixed depth (~ 1 μm). Therefore, a deposition process with excellent step coverage capability that can fill the space between actives without seam or void formation should be developed: this is one of the characteristics of the deposition process and is called “gap-fill characteristics.” In this regard, atomic layer deposition (ALD) has been employed as a gap-fill process using SiO₂ thin films. The ALD process exhibits self-limiting growth behavior induced by the chemisorption of a precursor at the surface of the substrate, resulting in a step coverage of over 95%. However, the excellent step coverage of the ALD process induces the formation of seams or voids. In the case of a high-aspect-ratio pattern with an ideal etch profile and a positive slope, the ALD process can fill the structure. However, the real etch profile of a high-aspect-ratio pattern would have a negative slope, resulting in tapering or bowing shapes owing to process difficulty. Where the sidewall of the trench or hole has a negative slope, the conformal SiO₂ film deposition induces the formation of seams or voids. In other words, the excellent step coverage characteristics of the deposition process hinder the gap-fill characteristics.

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Therefore, in the high-aspect-ratio pattern, higher growth rate at the bottom region of the trench than at the top region and surface would be favorable for filling the pattern without the formation of seams or voids. This is called "bottom-up growth". However, most of the deposition processes have a relatively higher growth rate in the top region of the trench ("top region") because the concentration of the reactant is higher in this region. Hence, the introduction of an inhibitor, which can suppress the growth of thin film deposition, was investigated to control the growth rate in this region. Among the various inhibition techniques, the plasma process is adequate for demonstrating the inhibition effect only in the top region, because the plasma generally has a large concentration gradient in the high-aspect-ratio pattern. However, plasma treatment can induce N or C contamination depending on the gas species, which can contribute to defects in the thin film.

Based on these considerations, the bottom-up growth of SiO₂ thin film deposition by employing a surface modification process via plasma treatment for the gap-fill process was investigated in this work. Plasma treatment was conducted by employing gases having a simple structure, namely, N₂ and NH₃, to avoid contamination. The inhibitory effect of the plasma treatment on the growth of the SiO₂ thin film was examined. Moreover, the mechanism involved in this inhibitory effect was evaluated. Eventually, the bottom-up growth behavior and gap-fill characteristics were investigated by employing plasma pre-treatment.

Results and discussion

Figure 1 and Table 1 present the growth inhibition effect of plasma pre-treatment depending on the gas species, namely, N₂ and NH₃. Before the plasma-enhanced ALD (PE-ALD) SiO₂ sequence was implemented, 1 s of plasma pre-treatment was performed for each gas species. The growth rate (growth per cycle, GPC) of PE-ALD SiO₂ was 0.064 nm/cycle without any plasma pre-treatment, and it decreased to 0.039 and 0.026 for N₂ plasma pre-treatment (N₂⁺) and NH₃ plasma pre-treatment (NH₃⁺), respectively. The decreased GPC ratios compared with the case of no inhibitor were 39.1, and 59.4% for N₂⁺ and NH₃⁺, respectively. The difference in the inhibition effect was attributed to the difference in the reactivity between N₂⁺ and NH₃⁺. As NH₃ consists of relatively weak chemical bonding, N–H, a relatively large number of radicals might be induced, resulting in higher growth inhibition in the PE-ALD SiO₂.

The chemical status and impurity incorporation during plasma pre-treatment were investigated using X-ray photoelectron spectroscopy (XPS). Figure 2a shows the depth profiles of deposited SiO₂ thin films with no treatment, N₂⁺, and NH₃⁺. In all cases, the deposited SiO₂ thin films had identical depth profiles for all elements, regardless of pre-treatment. To confirm the impurity incorporation by employing plasma pre-treatment, the depth profiles of C and N are depicted in Fig. 2b, which is a magnification of those in Fig. 2a. As shown in Fig. 2b, the concentrations of both C and N are less than 2 at.% which is the detection limit of the XPS measurement.
In addition, no increment in C or N concentration is observed by employing plasma pre-treatment. Because of the almost identical depth profiles and negligible C and N concentrations, plasma pre-treatment can inhibit the growth of SiO₂ PE-ALD without inducing any contamination or residue. Moreover, the stoichiometry of the deposited SiO₂ thin films was calculated (Fig. 2c). In the case of SiO₂ without pre-treatment, the O/Si ratio was 1.68 with homogeneity in the depth direction (standard deviation of 0.033). A relatively low O/Si ratio was attributed to the different sputtering yields between Si and O (lighter atom tends to easily sputtered out) during the etching for the depth profiling. The SiO₂ thin film with N₂⁺ had an O/Si ratio of 1.70 with a high uniformity (standard deviation of 0.017). In contrast, the O/Si ratio in the SiO₂ thin film with NH₃⁺ was relatively high at 1.78, with a slightly large fluctuation in the depth direction and a standard deviation of 0.075.

The chemical states of the deposited films were also investigated. Figure 3a shows the Si₂p XPS spectra of the SiO₂ thin films. All the spectra have identical profiles and only consist of Si–O bonding, corresponding to a binding energy of 103.8 eV\(^2\). However, in the O₁s XPS spectra, as shown in Fig. 3b, a difference is observed depending on the plasma pre-treatment. In the case of no inhibitor and N₂⁺, a shoulder at approximately 532 eV is observed, which is evidence that the peak at the lower binding energy contributed to the spectra. From the O₁s spectra, the composition of two bondings on the surface was evaluated (Fig. 3c) by the deconvolution to peaks at 532.6...
and 533.5 eV corresponding to the Si–O–Si and Si–OH bonding, respectively (Fig. 3d–f)²³. These two bonds are strongly related to the SiO₂ PE-ALD process. The Si–O–Si bond corresponds to the covalent bond of SiO₂. The Si–OH bonding is the surface termination of the SiO₂ thin film. In the ALD process, the precursor chemisorbs on the surface through a ligand exchange reaction with the functional group on the surface, which is called “anchoring site,” and only the chemisorbed precursor participates in the film formation reaction with the reactant, resulting in the self-limiting growth characteristics. The surface functional groups and their chemistry and density strongly influence the growth behavior in the ALD process. In the SiO₂ PE-ALD process, the Si–OH bonding, generally called the hydroxyl group, contributes to the ligand exchange reaction with disisopropylamino silane (DIPAS) (Si precursor)¹⁵,²⁴. The Si–OH bonding on the surface is changed to Si(from the surface)–O–Si(from the DIPAS)–H₃ during the precursor feeding step²⁴. After the reactant feeding step and the reactant purge step, the Si–OH bonding on the surface is recovered and participates in the chemisorption of DIPAS. Therefore, after the SiO₂ PE-ALD process is conducted, the surface consists of Si–OH bonds. In the case of SiO₂ deposition with NH₃*, the surface consists of only Si–OH bonding, implying that all the chemisorbed precursors fully react with the reactant and change into Si–OH bonding during the precursor feeding step²⁴. After the reactant feeding step and the reactant purge step, the Si–OH bonding on the surface is recovered and participates in the chemisorption of DIPAS. Therefore, after the SiO₂ PE-ALD process is conducted, the surface consists of Si–OH bonds. In the case of SiO₂ deposition with NH₃*, the surface consists of only Si–OH bonding, implying that all the chemisorbed precursors fully react with the reactant and change into Si–OH bonding. Moreover, the passivated Si–OH bonding during the NH₃* step recovers to Si–OH bonding during the SiO₂ PE-ALD process, resulting in growth inhibition without N contamination. In contrast to the case of NH₃*, the no inhibitor and N₂* cases exhibit Si–O–Si bonding ratios of 0.136 and 0.266, respectively. The Si–O–Si bonding on the surface might originate from the remaining Si(from the surface)–O–Si(from the DIPAS)–H₃ or the covalent bonding of SiO₂. Both cases indicate that the recovery of the Si–OH bonding during the reactant feeding and purge steps was suppressed or insufficient.

The gap-fill characteristics depending on the plasma pre-treatment were confirmed (Fig. 4). In the case of no inhibitor, voids in the trench are clearly observed, as shown in Fig. 4a. Although N₂* inhibits the growth of SiO₂, the void formed by the imperfection gap fill remains (Fig. 4b). Only in the case of NH₃*, a perfect gap-fill characteristic is obtained (Fig. 4c). Consequently, the inhibition of the growth of SiO₂ can enhance the gap-fill characteristics.

Further investigation of the growth inhibition effect by NH₃* was performed. First, growth inhibition depending on the ratio of NH₃* was evaluated. A treatment ratio of 0.1 indicates that the super-cycle consists of one cycle of NH₃* and nine cycles of SiO₂ PE-ALD (refer to Fig. 5). In the same manner, a treatment ratio of 0.5 means a super-cycle consists of one cycle of NH₃* and one cycle of SiO₂ PE-ALD. As shown in Fig. 6, the growth rate gradually decreases, displaying an almost linear relationship with an increasing treatment ratio, indicating that growth inhibition by employing NH₃* originated from the chemical inactivation of the Si–OH functional groups. The decreased Si–OH functional group density on the surface induces a decrease in the amount of chemisorbed Si precursor, resulting in a decrease in the growth rate. Moreover, the change in the growth rate depending on the NH₃ flow rate during NH₃* was examined (Fig. 7a). The NH₃ flow rate is related to the concentration of radicals formed in NH₃*. When the NH₃ flow rate was 50 standard cubic centimeter (sccm), the inhibition effect of NH₃* was slightly reduced from 0.026 nm/cycle to a growth rate of 0.031 nm/cycle. The decreased inhibition effect at an NH₃ flow rate of 50 sccm induced a deteriorated gap-fill characteristic, as shown by the void formation in Fig. 7b. As the NH₃ flow rate increasing to 100 sccm, the GPC was decreased to 0.026 nm/cycle, which is saturated value, and the seam-free gap-fill growth was observed (Fig. 7c,d). Furthermore, the time for NH₃* was varied at 500 sccm and the treatment ratio was 0.5. As shown in Fig. 7e, the growth rate increases from 0.026 nm/cycle to 0.36 and 0.45 nm/cycle as the treatment time decreases from 1.0 s to 0.5 and 0.25 s, respectively.
The growth rate of 0.45 nm/cycle at a treatment time of 0.25 s is comparable to that of 0.039 nm/cycle for \( \text{N}_2 \cdot \). This relatively high growth rate results in defective gap-fill characteristics (Fig. 7f). In the case of the treatment time of 0.5 and 1.0 s, the seam was not observed (Fig. 7g,h). In this regard, the difference in the stoichiometry (Fig. 2c) and surface termination can be ascribed to the change in the amount of the chemisorbed precursor. Even though the reactant was overdosed during the ALD process, the stoichiometry of the deposited thin film was strongly influenced by the density of the chemisorbed precursor on the substrate\(^2\). The higher O/Si ratio in the \( \text{SiO}_2 \) thin film prepared using \( \text{NH}_3 \cdot \) implies that the density of chemisorbed DIPAS decreased. Moreover, the recovery of surface termination to Si–OH (Fig. 3f) was facilitated by a relatively higher oxygen source ratio than the chemisorbed precursor.

These results confirmed that \( \text{NH}_3 \cdot \) can suppress the growth of \( \text{SiO}_2 \) thin films deposited via PE-ALD by inactivating the Si–OH functional groups on the surface. \( \text{NH}_3 \cdot \) induces bottom-up growth behavior during the \( \text{SiO}_2 \) PE-ALD process. In the case of no inhibitor (Fig. 8a–d), the introduced Si precursors were chemisorbed on the top and bottom of the trench-patterned structure (Fig. 8a) and exhibited the same growth rate. However, after \( \text{NH}_3 \cdot \) was performed (Fig. 8e–j), the inhibition only affected the top of the pattern (Fig. 8e), where the \( \text{NH}_3 \cdot \) was exposed; in turn, the growth was only inhibited on the surface and the top region (Fig. 8g). Therefore, the growth rate difference between the top and bottom regions of the pattern demonstrated bottom-up growth behavior (Fig. 8j). The inhibition of \( \text{SiO}_2 \) on the top region of the pattern by the \( \text{NH}_3 \cdot \) treatment was confirmed by performing a cross-sectional SEM analysis of the trench pattern (Fig. 9). For \( \text{NH}_3 \cdot \), the thickness of the deposited \( \text{SiO}_2 \) thin film on surface was 18.8 nm, whereas the thickness of \( \text{SiO}_2 \) deposited on the side was 32.5 nm. As the deposition was conducted, the bottom-up growth of \( \text{SiO}_2 \) in the trench pattern was observed. Furthermore, the bottom-up gap-fill characteristic was examined in a trench pattern with a small opening size and high aspect-ratio (opening and depth of 200 nm and 2.5 \( \mu \text{m} \), respectively) after conducting \( \text{SiO}_2 \) gap-fill using \( \text{NH}_3 \cdot \) by using the transmission electron microscopy (TEM) observation (Fig. 10). As shown in Fig. 10a, the trench was filled with \( \text{SiO}_2 \) from the bottom of the pattern to the top region of the pattern. The thicknesses
of SiO₂ on the side where the region not filled (Fig. 10b) was gradually decreased from the bottom of 89.5 nm to the top of 56.6 nm also indicating the bottom-up characteristic. Moreover, at the negative sloped area near the top area of the trench (Fig. 10c), the deposited SiO₂ film thickness gradually increased from the top (35.6 nm) to the bottom (58.5 nm). This is because of the relatively poor step coverage of NH₃ treatment. Due to the negative slope, the NH₃ was hard to reach on the surface of the side near the top area of the trench, inducing a gradient of the Si–OH functional groups on the surface. From the images of Fig. 10d,e, no seam was observed even in the TEM observation, indicating that the bottom-up gap-fill characteristics were achieved.

To confirm the seamless gap-fill characteristics more accurately, wet etching (using an etchant of diluted HF at a ratio of 200:1 for 60 s) was conducted, followed by cross-sectional SEM analyses. As shown in Fig. 11a, the seam propagates to the bottom of the pattern because of the infiltrated etchant in the case of the SiO₂ thin film deposited with no inhibitor. In contrast, in the case of NH₃, no seam is observed and only the surface is etched out with notch shapes (Fig. 11b). This profile indicates that no holes or voids are present where the etchant can infiltrate. In other words, perfect bottom-up growth was achieved by using NH₃. Owing to the bottom-up growth behavior, the gap-fill characteristics were significantly enhanced. The SiO₂ PE-ALD process with NH₃ was performed on the patterned structure, which had a varied trench opening size (Fig. 11c). On the large-opening-size trench, the trench was not fully filled, but it showed the bottom-up growth behavior; the level of the end point of the unfilled region (indicated by red circles in Fig. 11c) gradually moved upward to the surface. Moreover, bottom-up growth was observed even on the surface after completion of the gap fill of the trench (blue circles in Fig. 11c).

Conclusion
The bottom-up growth of the SiO₂ PE-ALD process was investigated by employing an inhibitor of NH₃ plasma pre-treatment to demonstrate seam- and void-less gap fill on an extremely high-aspect-ratio pattern. Plasma pre-treatment with N₂ or NH₃ gas effectively decreased the growth of SiO₂ PE-ALD without any contamination because of the suppression of the chemisorption of DIPAS on the substrate surface. The plasma concentration gradient in the trench structure induced a growth rate difference between beneath the trench and the surface, resulting in the relationship between enhanced gap-fill characteristics and the plasma pre-treatment process condition, which has a higher growth inhibitory effect. Owing to the inhibition effect of NH₃, the bottom-up growth behavior of SiO₂ PE-ALD was successfully implemented in the trench structure. Finally, a seamless gap-fill process was achieved in the high-aspect-ratio pattern.
Figure 8. Schematic diagram of the SiO₂ PE-ALD sequence consisting of (a) precursor feeding, (b) Ar purge, (c) O₂ plasma, and (d) Ar purge. Schematic diagram of SiO₂ PE-ALD using NH₃ sequence consisting of (e) NH₃ plasma, (f) Ar purge, (g) precursor feeding, (h) Ar purge, (i) O₂ plasma, and (j) Ar purge. To clearly describe the precursor chemisorption beneath the trench pattern and surface, the precursor chemisorbed on the sidewall of trench is omitted.
Figure 9. Cross-sectional FE-SEM images of the trench pattern (opening and depth of 150 and 400 nm, respectively) after conducting SiO₂ PE-ALD using NH₃ - for (a) 750, (b) 1000, (c) 1500, and (d) 2000 cycles.
Figure 10. (a) Cross-sectional TEM image of the trench patterns (opening and depth of 200 nm and 2.5 μm, respectively) after conducting the SiO₂ PE-ALD using NH₃⁺. (b) 20,000× magnified image of trench opening area, and (c–e) 80,000× magnified images of (c) trench opening area, (d) center of trench, and (e) bottom of trench (depicted as red boxes in (a)), respectively.

Figure 11. Cross-sectional FE-SEM images of the trench pattern (opening and depth of 200 nm and 2.5 μm, respectively) after conducting SiO₂ PE-ALD with (a) no inhibitor and (b) NH₃⁺, followed by wet etching (diluted HF of 200:1 for 60 s). (c) Cross-sectional FE-SEM images after conducting SiO₂ PE-ALD with NH₃⁺ on the trench pattern with a depth of 3 μm and an opening size varying from 300 (left) to 100 (right) nm.
Methods
SiO₂ thin films were deposited by applying PE-ALD(MAHA_AL, Wonik IPS) using DIPAS as the Si precursor and O₂ plasma with capacitively coupled plasma at a frequency of 13.56 MHz as the oxygen source. The Si precursor was introduced using a bypass-type source delivery system with a heated canister at 50 °C and an Ar flow as the carrier gas. The PE-ALD process temperature was 50 °C, and the sequence consisted of precursor feeding, Ar purging, O₂ plasma with a plasma power of 100 W, and Ar purging for 3, 5, 1, and 5 s.

Plasma pre-treatment was conducted prior to the SiO₂ PE-ALD sequence to inhibit the growth of SiO₂ PE-ALD. The plasma pre-treatment sequence consisted of gas feeding and plasma treatment, each for 1 s. Two types of gases, N₂ and NH₃, were employed, and the plasma pre-treatment was conducted at a process temperature of 50 °C with a plasma power of 100 W. The total deposition sequence is shown in Fig. 5.

The thicknesses of the deposited SiO₂ thin films were measured by applying ellipsometry (Aleris, KLA-Tencor). The depth profiles, compositions, and chemical states of the thin films were analyzed using XPS (NEXSA, ThermoFisher Scientific). The gap-fill characteristics of the SiO₂ PE-ALD process were examined by performing cross-sectional SEM (FE-SEM, JSM 760F, JEOL) and TEM (JEM-F200, JEOL) in trench structures having a depth of 1 μm and opening sizes varying from 100 to 300 nm.

Data availability
All data generated or analysed during this study are included in this published article.

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Author contributions
The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. Y.C., T.K., H.L., Jusung Park, and Juhwan Park performed thin film deposition, device fabrication, and characterization. D.R., and W.J. designed the experiment and co-wrote the paper. All of the authors discussed the results and commented on the paper.

Competing interests
The authors declare no competing interests.

Additional information
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