

# Ultrathin oxynitride films for CMOS technology

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**Abstract** — In this work, a review of possible methods of oxynitride film formation will be given. These are different combinations of methods applying high-temperature oxidation and nitridation, as well as ion implantation and deposition techniques. The layers obtained using these methods differ, among other aspects in: nitrogen content, its profile across the ultrathin layer, ... etc., which have considerable impact on device properties, such as leakage current, channel mobility, device stability and its reliability. Unlike high-temperature processes, which (understood as a single process step) usually do not allow the control of the nitrogen content at the silicon-oxynitride layer interface, different types of deposition techniques allow certain freedom in this respect. However, deposition techniques have been believed for many years not to be suitable for such a responsible task as the formation of gate dielectrics in MOS devices. Nowadays, this belief seems unjustified. On the contrary, these methods often allow the formation of the layers not only with a uniquely high content of nitrogen but also a very unusual nitrogen profile, both at exceptionally low temperatures. This advantage is invaluable in the times of tight restrictions imposed on the thermal budget (especially for high performance devices). Certain specific features of these methods also allow unique solutions in certain technologies (leading to simplifications of the manufacturing process and/or higher performance and reliability), such as dual gate technology for system-on-chip (SOC) manufacturing.

**Keywords** — *MOS technology, gate stack, ultrathin oxynitride layers, high temperature processing, plasma processing.*

## 1. Introduction

The ITRS Roadmap predicts that the successive technology nodes to be reached in the future still require aggressive scaling down of ultrathin dielectric layers serving as gate dielectrics in MOSFET's. It is commonly believed that as a result silicon dioxide (used since the beginning of the silicon technology era) will have to be abandoned, as the dielectric layers with higher dielectric constants would allow the use of thicker layers while preserving the gate capacitance.

It should be remembered, however, that there is yet another parameter that has to be taken into account while choosing the new gate-dielectric layer, namely the conduction band offset. Its value determines the current flow through the ultrathin dielectric layer (unless direct tunnelling currents dominate the leakage current): the lower the conduction band offset, the higher the current flowing through the MOS system.

A comparison of the materials considered as possible replacements for SiO<sub>2</sub> shows that the materials with dielectric constant tend to have lower values of conduction band offset (Fig. 1), which means that the choice is not obvious and some sort of compromise will have to be reached. Such difficulties will not occur if we stay with silicon dioxide and related compounds. In fact, careful studies prove that scaled-down pure silicon dioxide layers would perform well (in terms of leakage current) in high-performance devices, even down to the 22 nm node [1], and only low power and low standby power devices require serious technological changes.

Combinations of nitride and oxide layers have been successfully used for years in a number of applications; including even combinations of relatively thin but still separate layers, e.g. oxide-nitride (ON) or oxide-nitride-oxide (ONO) DRAM capacitor dielectric, or metal-nitride-oxide-silicon (MNOS) stacks for memory devices. In fact, an oxynitride layer is some sort of a curiosity, as it is not yet known at all why nitrogen atoms get incorporated into the oxide while nitride and oxide phases never coexist in the bulk under equilibrium conditions.

It was, therefore, obvious and tempting to investigate the possibility of the application of nitride and oxynitride layers as ultrathin gate dielectric in MOS devices.

In this thickness regime, no technology allows obtaining pure silicon nitride. In other words, some form of silicon oxide is always present. Thus, hereafter, we will use the terms "oxynitride layers" or SiO<sub>x</sub>N<sub>y</sub> for all ultrathin dielectrics discussed in this paper, keeping in mind that they may contain different amounts of silicon nitride.

The most important advantages expected of the application of oxynitride layers are as follows:

- much experience gained in the nitride/oxynitride processing during the years;
- compatibility of the material and its formation technologies with the standard silicon technology;
- high temperature stability;
- many possible techniques to choose from.

The aim of this work is to review the methods of the ultrathin oxynitride formation investigated so far and discuss their characteristic features, advantages and, therefore, the chances for application to ICs technology.

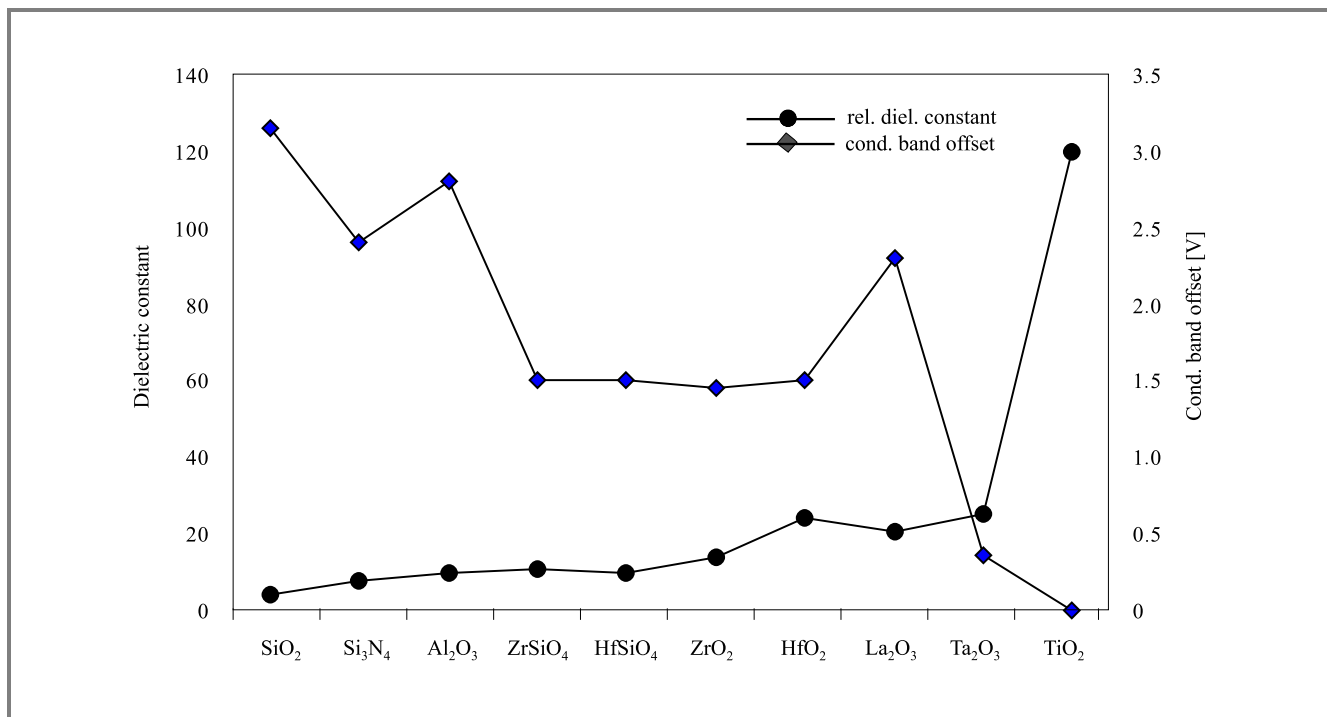


Fig. 1. Dielectric constant and conduction band offset of different dielectric materials.

## 2. “Ideal” ultrathin oxynitride layer

In most cases, the application of materials different than SiO<sub>2</sub> results in the deterioration of the silicon-insulator interface properties. The situation is, however, different in the case of the introduction of nitrogen into SiO<sub>2</sub> layers. It has been proved that the incorporation of some nitrogen into the silicon oxide gate dielectric can be beneficial for the technology and MOSFET device performance. Thus, ideal ultrathin oxynitride gate dielectric should exhibit:

- 1) nitrogen pile-up near or at the oxide-silicon interface – in order to improve the hot-electron immunity,
- 2) even higher nitrogen pile-up at the oxide-poly silicon interface – to suppress boron penetration from the poly-Si gate and to enhance device reliability.

It was also experimentally confirmed that the performance of MOS devices depends strongly on the concentration and the distribution of the nitrogen throughout the oxynitride layer (e.g. [2–5]).

Unfortunately, too much nitrogen at the interface is detrimental for MOSFETs’ performance, as excessive nitrogen at the oxide-silicon interface may lead to a reduction of the peak carrier mobility in the channel and allow boron accumulation in the oxide, leading even to device instabilities. Hence, many teams seem to prefer believing in the rule: “better no nitride pile-up at the interface, than too much”.

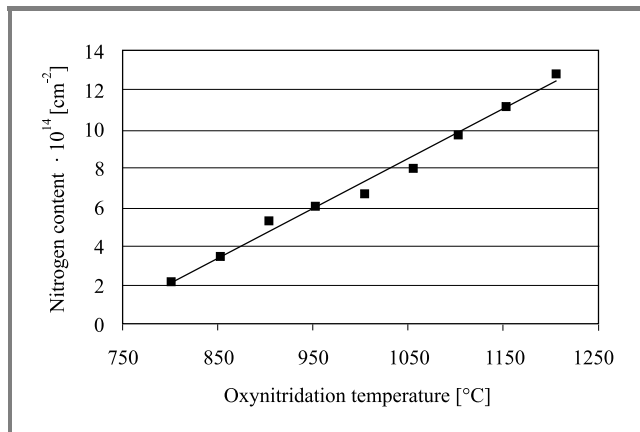
## 3. Oxynitride ultrathin layers formation

The list of the methods that could possibly be used to form ultrathin oxynitride layers is long. There are high-temperature methods, deposition methods (CVD or PVD) or even combinations of the two. Oxynitride layers may be produced in single process step but more often they are built up in two or even more steps in order to obtain the “ideal oxynitride layer”. These are:

- Thermal processes
  - silicon oxynitridation in NO,
  - silicon oxynitridation in N<sub>2</sub>O,
  - nitridation of previously grown oxide in NH<sub>3</sub>,
  - nitridation of previously grown oxide in N<sub>2</sub>.
- Deposition (CVD or PVD) and plasma assisted techniques
  - nitrogen implantation into silicon and then thermal or plasma oxidation (or the same in reverse order),
  - plasma nitridation in e.g. remote plasma,
  - PECVD deposition of an oxynitride layer,
  - atomic layer deposition (ALD),
  - jet vapour deposition (JVD).

### 3.1. Thermal processes

Thermal processes are performed at high temperatures not only due to the fact that the oxynitride-growth rate is relatively low when compared to that of the oxide, but also because of the observed dependence of the nitrogen content in the layer on the process temperature (the higher the temperature, the more nitrogen incorporated in the layer – see Fig. 2). Typically, however, the concentration of nitrogen (at the  $\text{SiO}_x\text{N}_y\text{-Si}$  interface) is of the order of  $10^{15} \text{ cm}^{-2}$ . When averaged across the whole layer it gives considerably less than 20% of the nitride in the oxynitride.



**Fig. 2.** Nitrogen content in 10 nm oxynitride films grown in  $\text{N}_2\text{O}$  by means of RTP (after [26]).

In order to obtain more nitrogen within the layer deposition methods have to be used. There are, however, several consequences of this change. First, low-temperature processing (deposition temperatures are usually  $< 400^\circ\text{C}$ ) results in a different structure of the layer (often non-equilibrium films). Moreover, (in contrast to high-temperature processes) a lot of hydrogen remains in the deposited layer. Both these features have much influence on the electrophysical properties of the whole gate system (silicon/oxynitride/poly-Si). This, in turn, may have considerable impact on the reliability.

Second, the layer is deposited onto the silicon surface, which is inevitably covered by a natural oxide. The thickness and nature of this oxide strongly depend on the method of chemical cleaning used. The oxide layer, rarely thinner than 0.4 nm, does not undergo much change during a low-temperature deposition process. Thus, unless special treatment (e.g. high temperature post deposition annealing) is used, it is precisely this layer that forms the silicon-oxynitride interface, which may obviously lead to poor interface properties of as-deposited layers. On the other hand, it may be expected that any high temperature process performed afterwards may considerably change the interface properties and composition profile of these layers.

As far as the formation of ultrathin oxynitride layers is concerned, high-temperature methods used to be the most obvi-

ous solution, especially since they have been used successfully for many years to form ultrathin oxide layers. Thus, they were also the first methods to be studied very carefully in the late nineties. The results were summarized in excellent reviews (e.g. [6, 7]).

There are important differences between depositing oxynitride layers in  $\text{NO}$  and  $\text{N}_2\text{O}$  ambients, although they might not be quite obvious at a glance. They result from the fact that the behaviour of  $\text{NO}$  and  $\text{N}_2\text{O}$  at high temperatures is different. The former is stable and diffuses to the interface, where the oxynitride is formed, while the latter rapidly decomposes and the reaction may follow many different paths (several tens of possible reactions), making the whole process difficult to control. Oxynitridation in  $\text{NO}$  is advantageous to process carried out in  $\text{N}_2\text{O}$  also due to the fact that it results in more nitrogen incorporated in the final layer (approximately  $< 15 \text{ at.}\%$  vs.  $< 5 \text{ at.}\%$  only for  $\text{NO}$  and  $\text{N}_2\text{O}$ , respectively). From the device point of view the advantages of oxynitridation in  $\text{NO}$  are lower leakage current and lower interface defect densities, as well as improved electrical stress properties. With a temperature rise, the concentration of incorporated nitrogen increases, and so does the width of the nitrogen containing region.

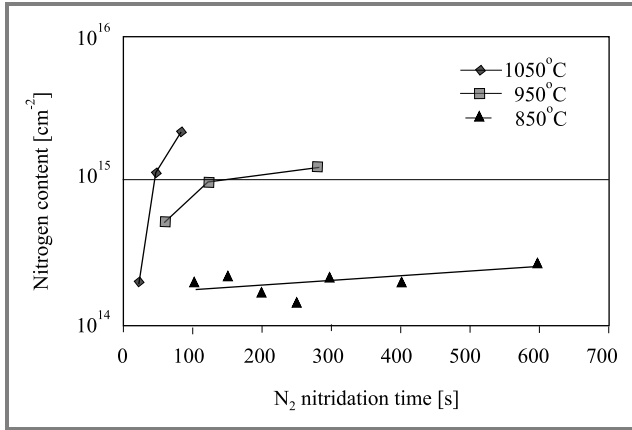
The kinetics of the growth of oxynitride films also make the process carried out in  $\text{NO}$  advantageous when compared to that performed in  $\text{N}_2\text{O}$ . In the former, the growth rate of the oxynitride film is much lower than in the latter. Thus, the process carried out in  $\text{NO}$  allows easier control (better process repeatability and layer uniformity) of the formation of extremely ultrathin oxynitride layers at high temperatures.

The differences in the nitrogen profile control are also observed between films grown in RTP (RTN) reactors and those grown in classical furnaces. The nitrogen profiles in the layers grown in the former are rather narrow and located at the interface, while in the latter much broader profiles are obtained. These effects are attributed to the location where the decomposition of the reactive gas takes place. In the case of furnaces it is the gas inlet of the hot wall tube, whereas in RTP reactive gas decomposes only in the vicinity of the hot wafer.

When nitridation of the previously grown ultrathin layer is considered, two types of ambient gas may be used, namely  $\text{N}_2$  or  $\text{NH}_3$ . Pure nitrogen is believed to be a relatively inert gas, but annealing of  $\text{SiO}_2$  layers in  $\text{N}_2$  has been proved (many years ago, already) to reduce the fixed charge in the oxide-silicon system, which has been attributed to the formation of a  $\text{SiO}_x\text{N}_y$  layer at the interface. Forming an oxynitride layer by means of a nitridation in  $\text{N}_2$  requires, however, a very high temperature – usually too high for the thermal budget typical of modern technologies. Replacing the furnace with an RTN may improve the situation a little, but not much (Fig. 3).

Nitridation of oxide layers in  $\text{NH}_3$  is definitely a more effective method of oxynitride formation. It allows incorporation of some 10–15 at.% of nitrogen into the layer. In

this case, however, the nitrogen piles up at both interfaces of the layer, namely: at silicon-oxynitride and oxynitride-poly-Si. Increasing the nitridation time results in more uniform nitrogen distribution, while almost no film growth is observed. The layers formed by this method exhibit, however, high concentrations of hydrogen that can play a role of traps. The concentration of hydrogen seems to follow



**Fig. 3.** Kinetics of nitrogen incorporation in an oxynitride layer during RTN (in N<sub>2</sub>) at different temperatures (after [27]).

that of nitrogen: the higher the temperature and the longer the process, the higher the concentration of both nitrogen and hydrogen. In order to reduce the hydrogen concentration within the layer, a high-temperature annealing process has to be performed.

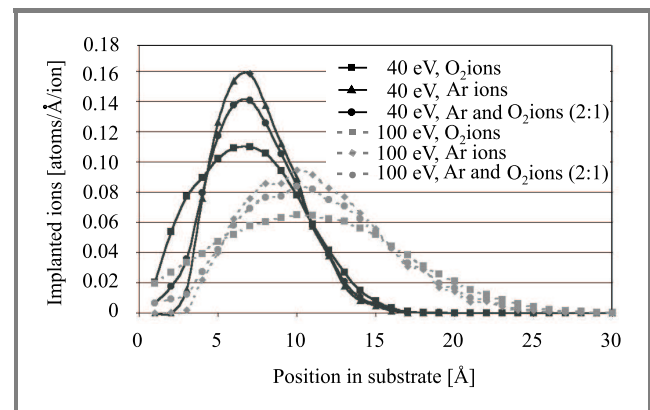
### 3.2. Deposition and plasma assisted techniques

A number of methods involving plasma or ion beam techniques (i.e. ion implantation) are studied for the application in oxynitride formation. Except for PECVD and JVD methods, where the oxynitride layers are formed in a single process, all the rest of these methods require a few processing steps (applying, except of ALD – different techniques).

Implantation of nitrogen atoms may be used to form a nitrogen rich layer in a silicon substrate, which then can be oxidised, either thermally or by means of a plasma-assisted method, to form the final oxynitride layer. This approach is particularly interesting for mixed IC or SOC technologies, where different gate oxide thicknesses are required depending on the role played by the individual devices (digital circuit, low power or memory). It has been shown (e.g. [8–10]) that the dose of the implanted nitrogen may be used to control the oxidation rate, and consequently, the final oxynitride thickness. This way, instead of individual processes of gate oxide formation for each type of devices, local implantations may be used to predetermine the final oxynitride thickness. Thus, oxynitride layers with different thickness are obtained in single a oxidation process (approach often referred to as “dual gate process”).

This sequence has some limitations that should be always kept in mind. Forming ultrathin oxynitride layers requires ultra-shallow implantation of nitrogen, which would suggest the use of very-low-energy implantation. Most implanters cannot, however, produce reasonable ion currents at low energies, which considerably reduces the process throughput. This is why attempts are being made to use alternative techniques, such as ion immersion implantation (III) (e.g. [11]) or even implantation from plasma sources, e.g. ECR (e.g. [12]), or r.f. plasma (e.g. [13]). These techniques lead to the formation of ultrathin oxynitride films with the peak of the nitrogen concentration located at the silicon-oxynitride interface (exactly as for all single-step high-temperature processes).

The reverse order of processing steps, i.e. oxidation first, nitrogen implantation later, may, on the other hand, offer some other technological possibilities. First, it is believed to result in lower concentration of defects in the oxynitride layer. This is most probably the consequence of the amorphization of silicon by implanted nitrogen, which does not get annealed out neither during the following low temperature plasma oxidation (temperature too low), nor even during a very short high-temperature oxidation (time too short). The effects of silicon surface damage due to ion bombardment during r.f. plasma exposure were studied e.g. in [13]. Even short exposure to low-power argon plasma in parallel symmetric electrode system, which is commonly believed to result in little radiation damage, appeared to increase considerably the oxidation rate of the subsequent plasma oxidation. This was attributed to the silicon amorphization during the exposure to argon plasma (see Fig. 4 where the probable distributions of very low energy argon and oxygen in silicon are shown).



**Fig. 4.** The distribution of Ar and O atoms implanted into Si substrate during plasma oxidation in a parallel plate symmetric reactor in pure O<sub>2</sub>, Ar and Ar:O<sub>2</sub> (2:1), as calculated with SRIM (after [28]).

On the other hand, even this low thermal budget annealing is enough to reduce considerably the initial nitrogen content in the film (especially during high temperature oxidation). The second approach enables also the silicon region to re-

main almost untouched, on condition that the implantation process is carried out very carefully (the right choice of energy and dose). This creates even more interest in very low-energy implantations performed from plasma sources (e.g. ECR). These processes yield totally different profiles of the nitrogen incorporation in the film – the maximum of the nitrogen profile stays away from the interface. As a consequence, the danger of carrier mobility degradation due to excess nitrogen at the interface is eliminated. In some extreme cases a pure- SiO<sub>2</sub> interface may remain, exactly as before the implantation process (e.g. [14]).

Plasma processes can be performed in different modes, such as planar reactors or remote plasma assisted process (RPAP), and using various plasma ignition methods, such as: microwaves, electron-cyclotron-resonance (ECR) or simply – radio-frequency (r.f.).

Among them, remote-plasma-assisted processing seems to have been studied particularly often in the past, e.g. [15–17]. Its potential advantages are:

- selective excitation of source and carrier gases (determined by their point of injection into the reactor chamber),
- substrate is located outside the plasma glow region,
- gas flow and pressure prevent backstreaming of source gases into the plasma generation region.

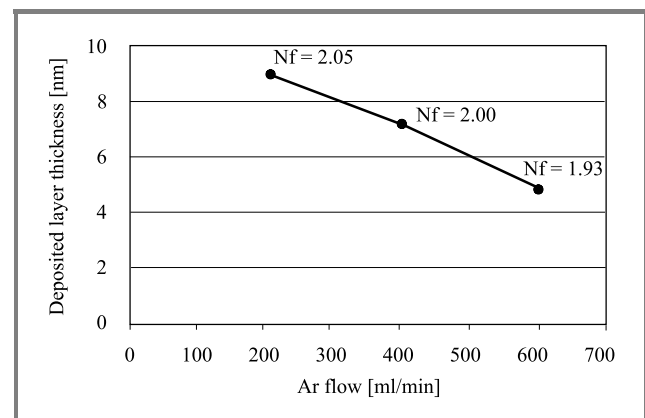
In this type of experimental set up two types of processes can be performed, namely: remote-plasma-oxidation (RPO) (or nitridation – RPN), or remote plasma enhanced chemical vapour deposition (RPECVD). In the first group, the active species react with the silicon substrate forming either oxide (RPAO) or nitride (RPN) layers. In the latter process type (RPECVD), the species activated in plasma undergo a chemical reaction and form oxide or nitride layers without consuming silicon atoms from the substrate, however, because these are supplied with the reactive gases (SiH<sub>4</sub> is typically used for this purpose).

The source gases used in the nitridation step are usually N<sub>2</sub>:He [16] or N<sub>2</sub>:Ar [16, 17], where both He and Ar particles serve as nitrogen excitation promoters. The resulting nitrogen concentration in the ultrathin oxynitride layer can be of the order of 8–10 at.% [17], which is similar to thermal nitridation in NO. It is, however, suggested that in order to obtain the best possible properties of the ultrathin oxynitride layer and, consequently, MOSFET devices, multi-step processing should be applied. An example of a set of 6 processing steps presented in [15] includes: surface etching, sacrificial oxide thermal oxidation, annealing, etching off sacrificial oxide, remote plasma oxidation, and remote plasma nitridation.

PECVD process allows basically a single-step deposition of an oxynitride layer. The main difficulty in this case is the reduction of the growth rate to the level allowing for good control of the ultrathin deposition process. The obvious move is to decrease the applied r.f. power and reactive-gas pressure, but in the case of an r.f. process a limit exists

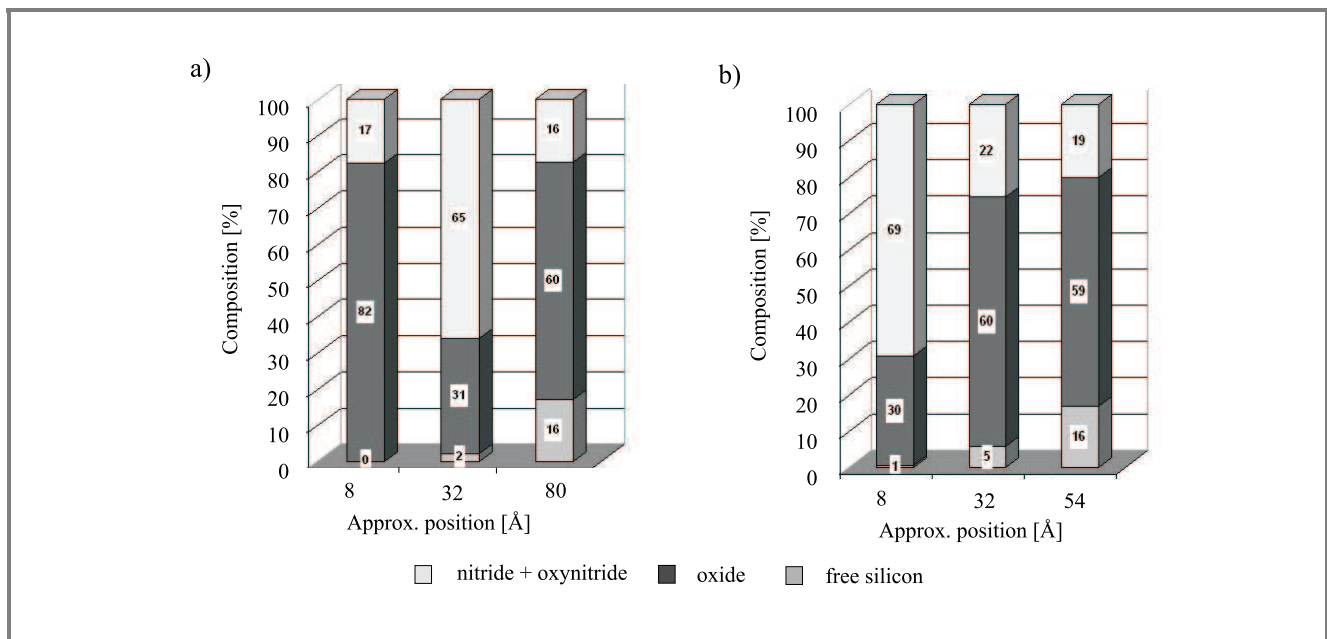
that results from the need to keep the plasma stable and uniform across the regions larger than the size of the silicon wafer.

Another possible solution to this problem is diluting the reactive gases in an inert gas, e.g. argon or helium. This can lead to precise control of the process within the whole ultrathin thickness range (Fig. 5) [18]. The bombardment of inert-gas ions can, however, also contribute to the final properties of the oxynitride layer. This effect is not always taken into account, as primary consequences of substrate amorphisation, i.e. higher oxidation (nitridation) rate, are difficult to distinguish from the dependence of the nitrogen excitation rate on the type of an inert gas used. In fact, it has been shown in [17] that even in remote-plasma reactors, believed to produce growth conditions almost free of ion bombardment, diluting N<sub>2</sub> with Ar may lead to significant changes not only in the deposition rate of the layer, but also in the nitrogen incorporation during the nitridation process. If other process conditions are the same, the nitrogen incorporation resulting from the use of N<sub>2</sub> or N<sub>2</sub>/Ar plasma is 8 at.% and 22 at.%, respectively. Other observations presented in this work prove that ion bombardment is essential to understand the process of plasma nitridation of oxide layers formed previously, because oxygen removal (e.g. by bombarding Ar ions) from the silicon oxide lattice seems to be a limiting step in the nitridation of such a film. Bombardment of the substrate with Ar ions during the deposition process (already mentioned above) leads to radiation damage in the deposited layers, that can be detected e.g. by means of XPS, as shown in [19].



**Fig. 5.** PECVD deposited nitride layer thickness as a function of argon flow. The process duration is 2 min. The respective refractive index values (@ 632 nm) are also shown (after [18]).

The primary difference between plasma deposition methods and the techniques of oxynitride formation discussed above is that they allow very high concentration of nitride (far beyond the reach of any other processes) to be obtained in oxynitride layers. Depending on the reactive gases used, the nitride concentration in an as-deposited ultrathin layer can be even as high as 95% and as low as 60%, or be-



**Fig. 6.** Comparison of XPS determined compositions at several selected positions within the layer for two different sets of PECVD parameters: (a) 83 Å layer (parameter set A) and (b) 60 Å layer (parameter set B) (after [20]).

tween 40% and 60%, as demonstrated by spectroscopic ellipsometry studies. Such high concentrations of nitride are often attributed to non-equilibrium, or nitrogen excess in the oxynitride structure.

On top of that, it has been shown that, in contrast to thermal processes, one can obtain different type of the nitrogen profile within the film by means of choosing appropriate PECVD process parameters [20]. In fact, as it can be seen in Fig. 6, PECVD appears to be very sensitive to process parameters in this respect.

Silane –  $\text{SiH}_4$  – is usually used as silicon source, although some papers report good results also with  $\text{Si}_2\text{F}_6$  or  $\text{SiH}_2\text{Cl}_2$ . In order to get high concentrations of nitride, ammonia ( $\text{NH}_3$ ) has to be used as the nitrogen source in plasma discharge, while for lower concentrations –  $\text{NH}_3$  and  $\text{N}_2\text{O}$ . Nitrogen, which is used for diluting  $\text{SiH}_4$  most often (in order to have a non-flammable mixture  $\text{N}_2$  98%:  $\text{SiH}_4$  2% is often used), is not effective in this respect (exactly as in the case of thermal processing). The use of ammonia in a low-temperature process has, however, another consequence, too. Oxynitride layers formed in this way exhibit unusually high concentration of hydrogen. Some reports claim even up to 30% of hydrogen in oxynitride layers. Due to the possible detrimental effects of hydrogen present in the dielectric-semiconductor system, a concentration of hydrogen this high cannot be tolerated. Thus, in order to improve the quality of the dielectric films in question, high-temperature annealing (usually RTA) is recommended.

Annealing of the ultrathin PECVD oxynitride layers at 600°C and 800°C in nitrogen was studied in [21, 22]. XPS studies, as well as optical and electrical characterization, have proved that the behaviour of the films is quite complex, as many effects take place simultaneously (mas-

sive hydrogen outdiffusion, film densification, nitrogen substitution by oxygen, etc.). Understanding these mechanisms still requires a lot of research to be done.

ALD is a type of chemical vapour deposition that typically allows deposition of less than a single monolayer per process cycle and has been successfully studied for a number of applications. Although the use of ALD to form ultrathin silicon-nitride gate-dielectric layers has been shown in a number of papers (e.g. [23, 24]), to our knowledge not much has been done so far in terms of identifying the obtained nitrogen profile or nitrogen content in the films. Thus, we will not comment on this method in this review.

The silicon nitride deposition by means of JVD is usually carried out in a co-axial dual-nozzle jet vapour source, in which highly diluted silane and  $\text{Ne}:\text{H}_2$  mixture flow into microwave discharge (e.g. [25]). Similarly to the case of ALD, most of the effort has been spent, so far, to prove the quality of the layer and its performance in the test devices and to our knowledge, no data on nitrogen content and profile are available.

## 4. Conclusions

Device requirements allow the concept of “ideal ultrathin oxynitride” gate dielectric film to be formulated. This concept should take advantage of both, the increase of hot-electron resistance caused by a limited (!!) nitrogen incorporation at the silicon-oxynitride interface, and the resistance to boron diffusion – due to a high content of nitrogen at the top of the dielectric film.

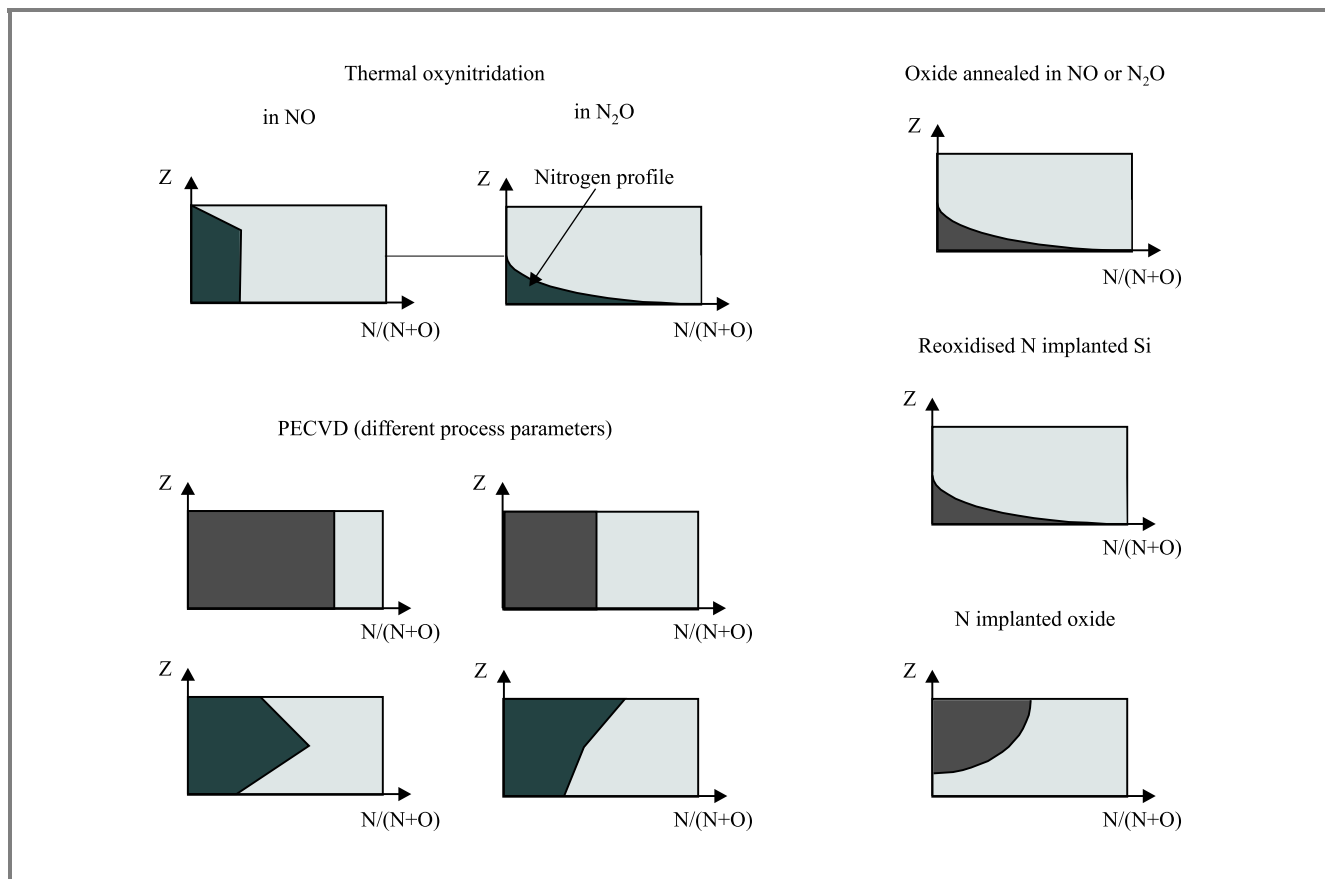


Fig. 7. Typical nitrogen incorporation profiles obtained by different technological methods of ultrathin oxynitride film formation.

In this review we have presented a variety of the methods that may be used to form ultrathin oxynitride and discussed nitrogen profiles obtained typically in the oxynitride layers formed (see Fig. 7). Thermal processes, so far studied the most heavily, do not provide nitrogen profiles of interest. The overall content of nitrogen in the layers formed in this way is lower, while the interface (in most cases) tends to be heavily nitrided. Multi-step processing has to be performed in order to overcome this limitation.

The methods involving ion implantation are more flexible in terms of the obtained nitrogen profiles. Due to the possibility of radiation damage, the technological difficulty is reducing the energy of ions reaching the sample, while simultaneously maintaining the control and the repeatability of the process. This group of methods is of particular interest due to the fact that oxynitride layers with different thickness may be manufactured in a single oxynitridation process – a feature exceptionally interesting for SOC technologies.

Growing oxynitride layers by means of PECVD appears the most flexible in this comparison. These methods may produce many different nitrogen profiles in the film in a single technological process, depending on the process parameters. On top of that, the incorporation of nitrogen in these layers can be the highest. Thus, even if it is remembered that the concentration of the nitrogen in the film is reduced

after high-temperature annealing, PECVD oxynitride layers still “hold the record”.

## 5. Acknowledgements

The authors would like to acknowledge also the contribution to this work of: T. Bieniek, M. Cuch and M. Giedz. This work was supported by research project no. 8T11B 074 19 financed by the State Committee for Scientific Research.

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