**Article**

**Influence of N<sub>2</sub> Flows on Sputtered Ta(N) films: Electrical, Structural, Chemical and Optical Properties**

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**Abstract:** By reactive DC magnetron sputtering from a pure Ta target onto silicon substrates, Ta(N) films were prepared with a different N<sub>2</sub> flow rate of 0, 12, 17, 25, 38, 58 sccm. The effects of N<sub>2</sub> flow rate on the electrical properties, crystal structure, elemental composition and optical properties of Ta(N) were studied. These properties were characterized by the four-probe method, X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), and spectroscopic ellipsometry (SE). Results show that the deposition rate decreases with an increase of the N<sub>2</sub> flows. On the other hand, the resistivity increases, the crystal size decreases, and the crystal structure transitions from β-Ta to TaN(111), and finally becomes the N-rich phase TaN(130,040). Studying the optical properties, it is found that there are differences in the refractive index (n) and extinction coefficient (k) of Ta(N) with different thicknesses and different N<sub>2</sub> flow rates, and dependent on the crystal size and crystal phase structure.

**Keywords:** tantalum nitride; electrical properties; structural properties; elemental composition; spectroscopic ellipsometry; optical properties

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1. **Introduction**

Transition metal nitrides, especially tantalum nitride (TaN) are highly demanded in a wide range of applications due to their high melting point, hardness, excellent wear and corrosion resistance, refractory character, mechanical and high-temperature stability, chemical inertness, and histocompatibility [1–6]. Some prominent examples of such applications as a protective coating material for protection against oxidation and corrosion [7], as a diffusion barrier for Al and Cu metallization in advanced microelectronics [8–11], in phosphide and nitride optoelectronics as ohmic contact [3,4], in artificial heart valves as histocompatibility materials [12], thin film resistor [13], as ceramic pressure sensors [14], and also different mechanical applications [5,6]. The large interest for TaN arises since it is considered recently as a high thermal conductive material in microelectronic chips for the β-TaN phase [15].

Furthermore, TaN belongs to the class of complex nitride phases of compounds which can vary from stoichiometry due to lattice vacancies. Diversifications in phase structure were common in TaN, from hexagonal-Ta₃N, cubic-TaN, hexagonal-Ta₅N, hexagonal-Ta₅N, tetragonal-Ta₃N and orthorhombic-Ta₅N including TaN, which led to large differences in physical, chemical, and mechanical properties. The TaN alloy forms a variety of phases depending on the deposition technique and growth conditions [16,17].
Numerous reports have been published to characterize sputtered TaN films based on various sputtering parameters such as nitrogen (N\textsubscript{2}) partial pressure ratio [18,19], N\textsubscript{2}/Ar flow rate ratio [20–22], sputtering power [23], and substrate temperature [24] during deposition. By controlling these different parameters, the influence on the structural, chemical, electrical, and optical properties of the TaN film have been investigated.

Among them, there are quite a few studies on how the N\textsubscript{2} flow or N\textsubscript{2}/Ar flow rate ratio and the N\textsubscript{2}/(N\textsubscript{2}+Ar) partial pressure ratio affect the properties of the TaN film. Chen et al. [25] used a magnetron sputtering low-power radio frequency deposition method with variable nitrogen flow rate to deposit TaN\textsubscript{x} barrier layers on silicon. They found that as the N\textsubscript{2} flow rate increases, the surface roughness of the deposited TaN\textsubscript{x} film was slightly increased, and the amorphous structure of TaN\textsubscript{x} was formed with good thermal stability. Zaman et al. [26] prepared a TaN film with a 3% to 25% N\textsubscript{2}/(N\textsubscript{2}+Ar) ratio on Si substrate by reactive magnetron sputtering and studied the effect of N\textsubscript{2} partial pressure on the crystal structure and hardness of the TaN film. It was found that the deposited films with 5% and 3% N\textsubscript{2} content showed the highest hardness (33 Pa).

Although sputtered TaN films have been widely investigated for their different properties, their optical properties have not been analyzed yet much. Recent studies have shown that spectroscopic ellipsometry (SE) can be used to characterize and measure the thin film thickness because of its fast and non-destructive nature [2,27–29]. Aouadi et al. [2] have studied the effects of varying N\textsubscript{2} flow rates from 1 to 4 sccm on the structural and optical properties of TaN\textsubscript{9} thin films. They report that the optical constants (ε\textsubscript{1}, ε\textsubscript{2}) will be used in conjunction with real-time SE to monitor and control the growth of tantalum nitride films. Cherfi et al. [30] deposited TaN films with an N\textsubscript{2} flow rate of 0-12 sccm on Si (100) and glass substrates by DC magnetron sputtering, and shown that the influence of nitrogen flow on the crystal structure and optical properties of TaN films. It was shown that for low N\textsubscript{2} flow (1 and 2 sccm), the TaN films show good conductor performance; a further increase of N\textsubscript{2} flow shows non-metallic behavior. At the same time, samples with similar structural properties have similar behaviors in terms of optical properties. Waechtler et al. have been shown that SE can be used to examine the optical properties of Ta and TaN thin films from 75 nm to 380 nm thickness. They have found a good agreement of optical properties with narrow-band data available for similar thin films. It was also shown that the optical properties of the films strongly depends with both substrate and film thickness [27]. Ma et al. have been studied the temperature-dependent dielectric function of TiN films by SE [28]. Recently, Xu et al. [31] used the method of comparing the measured refractive index of the low-k film under the Ta(N) diffusion barrier with the refractive index of the blank low-k film to study the integrity of the Ta(N) diffusion barrier using the approach developed by Shamiryan et al. [32].

However, there has been limited study of the optical behavior of the different stoichiometric of thicker TaN films by SE with varying N\textsubscript{2} flow rates in conjunction with electronic, structural and chemical composition. The study of the optical properties of TaN can provide us with more information about TaN films and some potential possibilities for the development of new applications. So, the systematic study of TaN films for understanding the electrical, structural, chemical composition and optical properties are required.

Therefore, we first focus our attention on the optical properties of TaN films by SE, a non-destructive testing method. By examining the optical properties of the sample, it can provide some guidance for the deposited sample. At the same time, from the existing research on the optical properties of TaN films, people ignore the influence of refractive index (n) and extinction coefficient (k), let alone explore the influence of process conditions on them, but they are also important optical parameters. Because the refractive index (n) and extinction coefficient (k) are regarded as “fingerprints of thin film materials.” Then, the effects of deposition rate and N\textsubscript{2} flow on Ta(N) films deposition on the electrical, structure, elemental composition and optical properties (n & k) of TaN films were studied by using the four-probe method, X-ray diffraction (XRD), X-ray photoelectron spec-
troscopy. The observation of different phases and chemical composition evaluation observed by XRD, XPS are correlated with optical properties.

2. Materials and Methods

Ta(N) films were synthesized using a standard magnetron sputtering (JS35-80G) system with sputtering non-uniformity is ≤±5%. The Ta(N) films were deposited on Si (100) wafers using magnetron sputtering of a Ta target (8.0 cm in diameter and 6.0 mm thick) of 99.95% purity. The substrate holder (located in the center of the chamber) was a 25 cm diameter plate, with rotation set to 10 rpm without heating the substrate for all of the depositions to improve the uniformity of the films. The target to substrate distance was 13.0 cm, and a negative bias was applied to the Ta target. After placing the Si(100) substrate in the deposition chamber, the chamber was evacuated to 9.6×10⁻⁴ Pa (by a turbo-molecular pump), the background vacuum was sufficient to ensure the vacuum required for Ta(N) film sputtering. Ar (99.999% purity) and N₂ (99.999% purity) were introduced into the reaction chamber through a mass flow controller and used as sputtering and reaction gases, respectively. The Ta target and the Si-substrate were sputter cleaned with Ar plasma prior to the Ta(N) films deposition for 5 min. Following cleaning, Ta(N) film was deposited at 9.56×10⁻⁴ Pa background pressure and 200 W DC applied power in a mixture of Ar and N₂.

To study the influence of different N₂:flow rates on the properties of the sputtered Ta(N) films, in all sputtering processes, the flow rate of Ar was fixed at 58 scm with various N₂:flow rates from 0 to 58 sccm. The ratio of the reactive gas (N₂, 0–58 sccm) to the sputter gas (argon, 58 sccm) were varied from 0, 0.20, 0.29, 0.43, 0.65, 1.00. The deposition time was also varied from 10 to 30 min to study whether the thickness of the film affects the optical and other properties of Ta(N) under each N₂:flow rate. Among them, when the N₂:flow is 12 sccm and 17 sccm, we only sputtered for 10 min respectively.

The thickness of the film was measured by a German Bruker Dektak step meter. The thickness was determined from the step height between the film and a masked substrate area. The resistivity of the sample was measured by a double-electric four-point resistance resistivity tester (FT-341) and was obtained from the current between two external probes and measuring the voltage through the internal probes.

The crystallographic structure of the sputtered Ta(N) films was measured by an x-ray diffractometer (Rigaku Ultima IV, Japan) using a 0–20 scan with a 1.54 Å wavelength Cu Ka radiation, at room temperature, working at 40 kV and 30 mA, and record the diffraction intensity in the scattering angle range of 20–60°.

X-ray photoelectron spectroscopy (XPS) was used to investigate the elemental composition and chemical states in Ta(N) films using PHI 5300 (PerkinElmer, USA) with an Mg Ka (1253.6 eV) excitation source. This source was operated using a voltage of 12.5 kV and an emission current of 20 mA. The films were sputter cleaned in an Ar⁺ environment with 89.45 eV pass energy for 5 min prior to measurement. Survey scans were conducted in the 0–1100 eV range.

The optical characterization of the films was carried out using a SENpro spectroscopic ellipsometer (SENTECH, Germany) to measure ellipse deflection phase (Δ) and amplitude (Ψ) angles at an incident angle of 70° in the spectral range from 400 nm to 1050 nm at 5 nm increments. In all cases, the ellipsometric data were processed using SpectraRay/3 software for the data analysis. By treating the Ψ−Δ spectra, the refractive index (n) and extinction coefficient (k) of the corresponding Ta(N) films were extracted by using E. Kondoh ELLIPSHEET [33] for an infinitely thick film. The validity of this approach will be proved below in the ellipsometry part of this paper. The specific details and film thickness are given in Table 1.
Table 1. The thickness, sheet resistance and resistivity of Ta(N) film deposited with different N₂ flow rates and different sputtering times.

| Film | Sputtering time (min) | Argon flow (sccm) | N₂ flow (sccm) | Thickness (nm) | Sheet resistance (Ω/sq) | Resistivity (µΩ.cm) |
|------|-----------------------|-------------------|----------------|----------------|-------------------------|-------------------|
| Ta   | 10                    | 58                | 0              | 223.18         | 8.67                    | 193.5             |
|      | 20                    |                   |                | 449.63         | 4.39                    | 197.4             |
|      | 30                    |                   |                | 664.35         | 2.92                    | 193.9             |
| TaN  | 10                    | 58                | 12             | 159.00         | 33.03                   | 524.00            |
|      | 10                    | 58                | 17             | 150.00         | 47.18                   | 738.00            |
|      | 20                    | 58                | 25             | 161.07         | 63.23                   | 1018.4            |
|      | 30                    | 58                | 28             | 282.08         | 55.57                   | 1567.5            |
|      | 10                    | 58                | 38             | 129.84         | 2090.0                  | 27136.6           |
|      | 20                    | 58                | 38             | 246.84         | 1020.0                  | 25177.7           |
|      | 30                    | 58                | 38             | 399.00         | 174.83                  | 6975.7            |
| TaN  | 10                    | 58                | 58             | 97.64          | 160.70                  | 1568.9            |
|      | 20                    | 58                | 58             | 195.93         | 7480                    | 146555.6          |
|      | 30                    | 58                | 58             | 305.19         | 13200                   | 402850.8          |

3. Results and discussion

3.1. Deposition rate and resistivity

The deposition rate of Ta(N) films, sheet resistance and resistivity depend on the flow of nitrogen. The film thickness decreases with increasing the flow of nitrogen, as shown in Figure 1a and the increase of sputtering time from 10 min to 30 min makes the film thicker. However, the deposition rate does not depend on the sputtering time (Figure 1b). With the increase of nitrogen flow in the sputtering atmosphere of ionized Ar⁺, the intensity of ion bombardment of Ta target decreases due to the reduction of mean free path length and the number of sputtered Ta atoms also decreases leading to a gradual reduction in deposition rate [34]. In addition, since there are a large number of active N atoms in the sputtering atmosphere (an increase of the N₂ flow rate), the number of active N atoms in the atmosphere gradually increases, which increases the chemical reaction between active N atoms and the surface of the Ta target. The probability of TaN compound causes slight poisoning of the target [35,36], thereby reducing the sputtering rate.
Figure 1. Change of Ta(N) film thickness measured by Dektak step meter (a) and the deposition rates (b) for different nitrogen flows and sputtering time.

The electrical resistivity of pure Ta films sputtered for 10, 20 and 30 min are nearly same (193.5, 197.4, and 193.9 \( \mu \Omega \text{-cm} \), respectively (Table 1)). It is interesting to notice that the measured resistivity for pure Ta films are similar to the values reported for tetragonal crystalline Ta (\( \beta\)-Ta) films (165 \( \mu \Omega \text{-cm} \), Schauer et al. [37]; 210 \( \mu \Omega \text{-cm} \), Cuong et al. [38]) and 242 \( \mu \Omega \text{-cm} \), Arshi et al. [39]). Therefore, our pure Ta films are most likely \( \beta \)-Ta. The electrical resistivity of Ta(N) films deposited with different nitrogen flows is shown in Figure 2. It can be seen that the resistivity of TaN is higher than the resistivity of pure Ta observed at zero nitrogen flow. Introduction of nitrogen increases resistivity: first, it changes linearly (embedded graph) and thinner film has lower resistivity. The trend of increased resistivity can be attributed to the decreasing of the low resistivity Ta phase in the deposited Ta-N films and to the increasing of the low resistivity N-rich TaN phase. A much more dramatic change of resistivity is observed in the films deposited with 58 sccm N\(_2\) flow, especially after 30 min deposition (relatively thick films, see Table 1).

Figure 2. The resistivity TaN films at different nitrogen flow. The embedded graph is a zoom of data corresponding to low nitrogen flow region.

Further increase of N\(_2\) flow increases the resistivity of Ta(N) film and might depend on the formation of different surface topography, grain size, changes of composition, amorphous structure formation and defects/imperfection (scattering from grain boundary) [40]. The resistivity of TaN films in the current work is similar to the values observed in the literature [40,41]. When the N\(_2\) flow rate is 12 sccm, the resistivity of the sputtered TaN film is increased to 524 \( \mu \Omega \text{-cm} \) and close to FCC TaN [42–45], or cubic TaN(111) [39] or Ta\(_3\)N\(_5\) or Ta\(_5\)N\(_6\) [46–48]. After a further increase in the nitrogen flow to 25 sccm, the resistivity of the sputtered TaN film is close to the resistivity of Ta\(_3\)N\(_5\)(1126 \( \mu \Omega \text{-cm} \), [39]). Remarkably, when the increase of N\(_2\) flow was increased to 38 and 58 sccm, the resistivity of the sputtered TaN films drastically increased, with only exception of 10 min deposition TaN films. Normally, it can be explained by increasing electron scattering from interstitial N atoms. [39]. However, this model does not explain so strong difference between the films deposited with 38 and 58 sccm [49]. It is also well-known that an excess of N\(_2\) flow rate will decrease the mean free path of ionized Ta atoms and disturb the formation of TaN structures [40,50] and also increased electron scattering from interstitial N atoms. Therefore, the phase of the TaN film generated under our N\(_2\) flow rate will also be dif-
different. The existence of N-rich phases in the TaN films at higher nitrogen flows is consistent with both the XRD patterns and the XPS analysis.

3.2. Structural Properties (XRD Analysis)

Figure 3 shows the X-ray diffraction (XRD) patterns of the Ta(N) films deposited for 10 min, 20 min, and 30 min with different N\textsubscript{2} flows in the gas mixture varying from 0 to 58 sccm. The XRD spectra of the Ta films (N\textsubscript{2} flow rate is 0) shows a mixed phase of β-Ta (221), β-Ta (002), β-Ta (330), and Ta(110), among which β-Ta The diffraction intensity of (002) is the highest, and the peak area of the diffraction peak is the largest, which indicates that the Ta film we sputtered is mainly β-Ta (002) preferred orientation (PDF#: 04-0788). Peaks at 35.40°, 37.04° and 41.20° are indexed to be the TaN (111), TaN (111) [39], and TaN (200) (PDF#: 49-1283) structure respectively (Figure 3a). The peaks at 31.86°, and 35.10°corresponds to TaN\textsubscript{s} (123) and TaN\textsubscript{s} (130) or (040) compounds respectively [51].

When the N\textsubscript{2} flow rate is 12 and 17 sccm, a mixed phase of TaN (111) and TaN (200) appear, and the diffraction peak of TaN\textsubscript{s} (023) also appears when the N\textsubscript{2} flow rate is 17 sccm. However, the diffraction peak of TaN (111) is the highest, and the diffraction peak area is also the largest, which indicates that the preferred orientation of the TaN film under these two flow rates is TaN (111). Similarly, when the N\textsubscript{2} flow rate is 38 and 58 sccm, the TaN film has a preferred orientation of TaN\textsubscript{s} (130,040). At the same time, when the N\textsubscript{2} flow rate increases, the diffraction peaks gradually widen, which indicates that the grain size is gradually decreasing that leads to high resistivity Table 1 and correspondingly it could be attributed to the mixture of fcc TaN and amorphous structure [46,52,53]. These phenomena observed by XRD are in line with our previous conjectures in the section on resistivity and similar to those reported in an earlier study [43,44].

When the nitrogen flow rate is increased, the phase of the film evolved gradually from TaN(111) to TaN\textsubscript{s} (130) or TaN\textsubscript{s} (040) (35.40° to 35.03°) [51]. On the other hand, the TaN (200) peaks are gradually decreasing. A broad peak corresponding to TaN\textsubscript{s} appeared for the 58 sccm sample and significant broadening of the peaks could be due to the formation of a two-phase nanocomposite structure. This can be attributed to the high nitrogen fraction, which is known to inhibit the crystallization of nitrogen rich TaN\textsubscript{s} sputtered films. At the same time, the XRD patterns of TaN films with a nitrogen flow rate of 25–58 sccm and a sputtering time of 10–30 min were compared (Figure 3b and 3c), and it was found that the XRD patterns of TaN films with the same nitrogen flow rate but different sputtering times/different thicknesses did not change much, which shows that the thickness of the film does not affect the formation of the crystal structure of the film. Generally, the films deposited with 58 sccm of nitrogen do not have pronounced patterns and this suggests that the films are losing their crystalline structure and becoming more amorphous (Table 2).

Table 2. Calculated crystallite sizes of Ta and TaN films.

| Flow rate of N\textsubscript{2} (sccm) | Peaks | Peak position, 2θ (deg) | FWHM, θ (deg) | Average crystalline size (nm) |
|-----------------------------------|-------|------------------------|---------------|-------------------------------|
| 0                                 | β-Ta (002) | 33.72                  | 0.266          | 31.22                         |
| 12                                | TaN(111)  | 35.40                  | 0.340          | 24.54                         |
| 17                                | TaN(111)  | 35.28                  | 0.394          | 21.15                         |
| 25                                | TaN_s (130, 040) | 35.04                  | 0.543          | 15.34                         |
| 38                                | TaN_s (130, 040) | 35.12                  | 0.890          | 9.37                          |
| 58                                | TaN_s (130, 040) | 34.24                  | 3.394          | 2.45                          |
Figure 3. XRD patterns of Ta (20 min) and TaN films deposited for (a) 10 min, (b) 20 min, and (c) 30 min with different N₂ flows.

3.3. Elemental composition (XPS Studies)

XPS spectra was obtained to ascertain the elemental composition of the deposited TaN films. Figure 4 illustrates an evolution of the XPS survey spectrum of the deposited TaN films as a function of N₂ flow rate in the binding energy range of 0–1150 eV and show the Ta, O, N and C signals. It is confirmed that the survey does not contain additional component arises in the Si₂p spectrum because our films were thicker. The unexpected O and C signal in these spectra might come from the ambient atmosphere in the sputtering chamber and/or the presence of background oxygen in the chamber during sputtering, and/or from organic residues during the storage, as already reported in literatures [35,39,54], also depends on different sputtering instruments, although there is a chemical shift for both O and C in the lower binding energy with increasing the N₂ flow rates due to the formation of Ta₂O₅ (Figure 5a and 5b).
Figure 4. Evolution of the XPS survey spectra in the binding energy range of 0–1100 eV as a function of N₂ flows (12–58 sccm, 10 min deposition) showing the Ta, O, N and C signals for TaN films.

Figure 5. XPS core levels spectra of (a) O1s, and (b) C1s for the TaN films at different N₂ flows (12–58 sccm, 10 min deposition).
The XPS core-level spectra of Ta4f, Ta4d and N1s for different N2 flow rates (12–58 sccm) were shown in Figure 6a, Figure 6b and Figure 6c, respectively. Figure 6a shows the XPS region of Ta4f, revealing that it is composed of three overlaying bonding environments: Ta4f{5/2} of Ta-O (Ta bonded with O), Ta4f{7/2, 5/2} of Ta-N and Ta4f{7/2} of Ta-N located at 30.25, 28.15, and 25.97 eV, respectively. As shown in Figure 6a, increasing the flow of N2 from 12 to 58 sccm is likely to chemical shift the Ta4f{7/2} peaks from 25.97 to 25.78 eV, which were attributed to TaN Ta4f{5/2} peaks. On the other hand, the chemical shift from 30.25 to 29.81 revealing that O-rich films composed of Ta2O5 with an increase of N2 flows were observed. Figure 7 shows the deconvoluted spectra of Ta4f for the TaN films with N2 flow of 12 sccm. Compared with the Ta binding energy values of TaN and TaOx in references, the binding energy values in the Ta4f spectrum (Ta4f{5/2} = 25.7 eV and Ta4f{7/2} = 27.7 eV) accorded with the chemical state of Ta in Ta–O binding [55]. The Ta4f doublet at binding energy Ta4f{7/2} = 27.3 eV and Ta4f{5/2} = 29.0 eV matched Ta5+ state in Ta2O5 [56,57], while the corresponding Ta4f doublet peaks are located respectively at Ta4f{7/2} = ~25.1 eV and Ta4f{5/2} = ~27.3 eV that should be attributed to N-rich TaN phase [58,59].

Figure 6. XPS core levels spectra of (a) Ta4f, (b) Ta4d and (c) N1s for the TaN films at different N2 flows (12–58 sccm, 10 min deposition).
Figure 7. Deconvoluted spectra of Ta4f for the TaN films with N2 flow of 12 sccm for 10 min deposition.

The existence of the Ta–N bonding in the film is further confirmed by the N1s peak located around 397.0 eV [60]. As the nitrogen flow rate increases, more Ta–N bonds form and the N1s peak also increases. This is consistent with the XRD spectra. It can be seen from Figure 6c that the N1s peak shifts from 397.55 to 398.85 eV for nitrogen flow from 12 to 58 sccm, while the Ta4p3/2 peak also shifts from 406.4 to 404.95 eV. It is noteworthy that there are uncertainties on the exact value for Ta4p binding energy. Nyholm et al. reported a Ta4p value of 400.9 eV [61] and Tian et al. [62] reported a relatively high value of 406.0 eV. This shift in the binding energy for the N1s peaks toward lower values is in agreement with a previous study [35,39], while the binding energies for the Ta4f doublets also shift toward lower energy values and do not agree with previous studies.

3.3. Optical Properties

Spectroscopy ellipsometry (SE) is broadly used as an important tool for thin films thickness, refractive indices and optical properties analysis, and its basic principle is shown elsewhere [28]. For optically thick films, SE spectra can be fitted using a single layer (substrate model) and optical properties of the films can further be analyzed. As the refractive index (n) and extinction coefficient (k) cannot be measured directly and so must be calculated by some quantities that are related to them and can be directly measured. In this study, ellipsometry parameters Ψ and Δ were obtained by SE with Spectrumfit Levenberg-Marquardt + thickness scan fitting. Therefore, we measured the Ψ and Δ of all Ta films and TaN films by using only the substrate model, and extracted the n and k of the corresponding film by using E. Kondoh ELLIPSHEET [33].

Figures 8a and 8b depict the complex refractive index (n) and extinction coefficient (k), respectively for Ta films and with a comparison of the literature report. After comparing with the data of Tompkins et al. [63]. and Waechtler et al. [27], it is found that the changing trends of n and k of Ta films with different thicknesses are roughly the same, and the thickness does have an effect on the optical properties of Ta films. It is different from the conclusion that Waechtler et al. reported the same n, k of Ta films with different thicknesses.
Figure 8. Refractive index (n) and extinction coefficient (k) of Ta films sputtered for 20 min (450 nm) and 30 min (664 nm) are compared with literature data.

Figure 9. The refractive index (n) and extinction coefficient (k) of TaN(12-58 sccm) films sputtered for 10 min are compared with those in the literature.

In Figure 9, we compare the refractive index (n) and extinction coefficient (k) of our sputtered TaN film with an N₂ flow rate of 12-58 sccm with the data of Aouadi et al.
al. [2] and Waechtler et al. [27]. Also, it is found that when the N$_2$ flow rate is 12 and 17 sccm, the n and k of our sputtered TaN film are close to the values in the reference, but as the N$_2$ flow rate increases, the n and k values of our sputtered TaN film are getting less and less close to the n and k values in the references (Fig. 9a and b). Besides, different N$_2$ flow rates and different thicknesses of TaN films have significant differences in n and k, which shows that both N$_2$ flow rate and thickness affect the optical properties of TaN. In addition, we can see that there are differences in the n and k values and curve shapes of TaN films with different crystal structures. The influence of N$_2$ flow rate on the n and k of TaN film may be caused by the different crystal structures and grain sizes of TaN film deposited with different N$_2$ flow rates. It is interesting that the extinction coefficient of TaN films deposited at high nitrogen flows (38 and 58 sccm) decreases starting from 700 nm and especially from 900 nm. The reduction of extinction coefficient suggests that the films are becoming more like dielectric and explains the drastic increase of their resistivity (Figure 2).

In this work, we used optically thick films to be able to extract optical characteristics. The thickness of these films are measured by using a Bruker Dektak step meter that also is used only for relatively thick film. However, many applications of Ta and TaN layers need very low thickness. For instance, when they are used as diffusion barriers in advanced microelectronics. Taking it into account we examined the applicability of ellipsometry to measure the thickness of the thin Ta and TaN layers. For this purpose, we used values of optical characteristics of these layers found in literature and measured in our work. Then we calculated $\Delta-\Psi$ trajectories for the films with the different thickness (Figure 10). The presented curves demonstrate that Ta(N) films thickness can be measured by ellipsometry when d $\leq$ 100 nm. The sensitivity is reduced with thickness but it can be very good for evaluation of Ta(N) films deposited as diffusion barriers for microelectronics application (d < 10 nm). It is also obvious that ellipsometry will be efficient for evaluation of the d < 100 nm films continuity as it was done in the Ref. [31,32].

4. Conclusions

The Ta(N) film with different N$_2$ flow rate (0-58sccm) and sputtering time of 10-30 min was deposited by the DC reactive magnetron sputtering method, and it was found that the deposition rate Ta(N) film, electrical, structural, chemical and
optical properties depend on N₂ flow rate. As the N₂ flow rate increases from 0 to 58 sccm, the crystal structure of the sputtered film transitions from β-Ta to TaN(111) and finally becomes the N-rich phase Ta₃N₅ (130) or TaN₅ (040). When the N₂ flow rate increases, the diffraction peaks gradually widen, which indicates that the grain size is decreasing that’s leads to higher resistivity (Table 1) and correspondingly it could be attributed to the mixture of fcc TaN and amorphous structure The films deposited with 58 sccm of nitrogen lose specific crystallographic patterns and therefore becoming amorphous. In the part of the optical properties study, we can see that both the thickness and the N₂ flow rate will affect the refractive index (n) and extinction coefficient (k) of TaN film, and have a greater impact on k. The curve shapes of n and k of similar crystal structures have a small difference. The influence of N₂ flow rate on the refractive index and extinction coefficient of TaN film may be caused by the different crystal structures and grain size of TaN film deposited with different N₂ flow rates. The extinction coefficient of the films deposited with 58 sccm of nitrogen decreases that suggests the formation of a more dielectric like nature of the deposited films. This fact explains the drastic increase the resistivity of the films shown in Figure 2. The reason for the transformation to the dielectric state is the incorporation of Ta oxide that can be seen from XPS data. When the nitrogen flow is so large, it reduces the free path length of sputtered atoms, poisons the Ta target and therefore reduces the effective deposition rate. As a result, the role of the residual oxygen is drastically increased and the deposited film is becoming Tantalum oxynitride with much higher resistivity. This effect is becoming more pronounced when the deposition time is long and this is the reason of the strong difference in resistivity between the films deposited during 10 min and 30 min. If deposition time is short, the target poisoning might be negligible.

Finally, the presented results suggest the resistivity of TaN films deposited by magnetron sputtering can be precisely controlled by changing nitrogen concentration during deposition. It is also important that a too high concentration of nitrogen might have a negative effect increasing the impact of gas phase impurities.

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