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Short Communication

Silicon nitride thin-films deposited by radiofrequency reactive sputtering: Refractive index optimization with substrate cooling in a nitrogen-rich atmosphere

João R. Freitas [a,*], Sara Pimenta [a,b], Vítor H. Rodrigues [a], Manuel F. Silva [a,b], José H. Correia [a,b]

[a] CMEMS-UMinho, University of Minho, 4800-058, Guimarães, Portugal
[b] LABBELS-Associate Laboratory, Braga/Guimarães, Portugal

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ABSTRACT

Silicon nitride (SiN) is widely used as a core material in optical waveguides due to its optical properties. The deposition of SiN thin-films by radiofrequency (RF) reactive sputtering is commonly used in low-temperature processes, where the thin-films optical properties can be optimized by controlling the deposition parameters (sputtering power, gases ratio, etc.). This work presents the deposition of several SiN thin-films by RF reactive sputtering with different sputtering powers (ranging from 180 W to 300 W), with a nitrogen-argon ratio of 16:4, and performing substrate cooling in a nitrogen-rich atmosphere immediately after deposition, consisting in keeping the substrate under 16 sccm of nitrogen until it reaches 25 °C. The refractive indices of the SiN thin-films were assessed through ellipsometry, obtaining a maximum refractive index of 1.906 at 400 nm. SiN thin-films were also analyzed by energy-dispersive spectroscopy (EDS) and atomic force microscopy (AFM).

1. Introduction

In recent years, silicon nitride (SiN) has gained much attention as a core material for optical waveguides in near-infrared and visible spectral regions [1–3]. SiN has a relatively high refractive index (≈2), confining the light inside the waveguide core. SiN thin-films are commonly deposited by chemical vapor deposition (CVD) techniques, i.e., low--pressure CVD (LP-CVD) or plasma-enhanced CVD (PE-CVD) [4,5]. However, for low-temperature processes (e.g., polymeric-based devices) and low hydrogen (H) content in the films, physical vapor deposition (PVD) of SiN is widely used. This can be achieved by using techniques such as reactive magnetron sputtering [6–8], dual ion-beam sputtering (DIBS) [9,10], and ion-assisted deposition (IAD) [11–13].

Radiofrequency (RF) sputtering is a physical vapor deposition process that occurs in an evacuated chamber, with a low pressure of an inert gas (argon (Ar)). This process uses a high voltage alternating current power source that generates energetic waves, ionizing the Ar gas. The high-energy Ar^+ ions collide with a target material, sputtering off its atoms that will deposit on a substrate [14,15]. Varying the sputtering deposition parameters makes it possible to change the final optical and mechanical properties of the SiN thin-films [16–18].

RF reactive sputtering of SiN thin-films is commonly performed using a silicon (Si) target and a nitrogen-argon plasma. Several authors have reported the tunning of SiN refractive index by changing several deposition parameters, observing an increase in the SiN refractive index with the increase of sputtering power, substrate heating during deposition, high nitrogen-argon ratio during deposition, and lower sputtering pressure [8,19–22].

In the present study, several SiN thin-films were deposited by RF reactive sputtering under different sputtering powers, with a high nitrogen-argon ratio, and performing substrate cooling in a nitrogen-rich atmosphere immediately after deposition, consisting in keeping the substrate under 16 sccm of nitrogen until it reaches 25 °C. The SiN thin-films were assessed through different methods. The refractive indices were obtained through ellipsometry, the element composition was analyzed by energy-dispersive spectroscopy (EDS), and the surface topography was acquired through atomic force microscopy (AFM).

2. Experimental details

SiN thin-films were deposited from a Si target (Intrinsic Si, *Siltronix*), using RF reactive sputtering.

Firstly, as an initial test, SiN thin-films were sputtered on a silicon substrate with 3 different power supplies (100 W, 150 W, and 200 W),

* Corresponding author.
E-mail address: id9493@uminho.pt (J.R. Freitas).

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Table 1
Deposition parameters of SiN thin-films by RF reactive sputtering.

Power (W)	Nitrogen-argon ratio	Pressure (mbar)	Deposition rate (Å/s)	Substrate cooling in a nitrogen-rich atmosphere
100	13:7	6×10^{-3}	0.4	No
150			0.6	
200			1	
180	16:4	6×10^{-3}	0.7	Yes
200			0.8	
230			0.8	
250			0.8	
300			0.9	

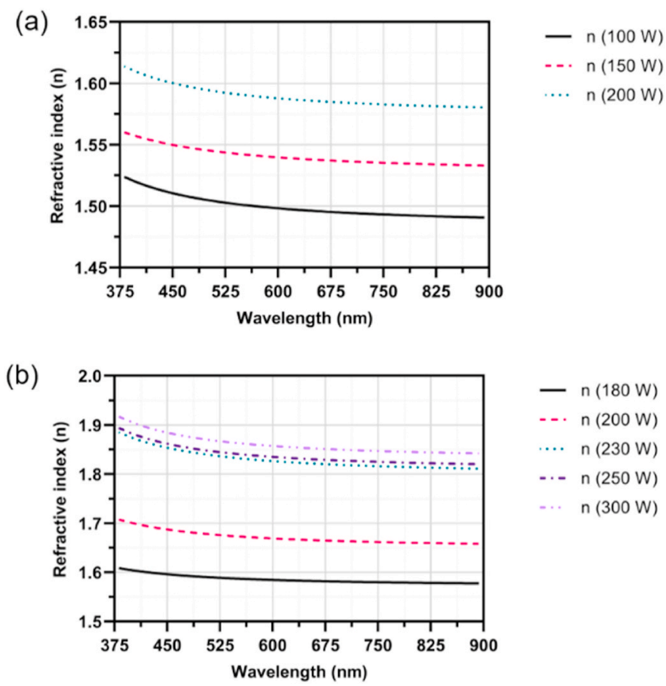

Fig. 1. Refractive index curves as a function of the wavelength obtained in several SiN thin-films: (a) sputtering power between 100 W and 200 W, nitrogen-argon ratio of 13:7, and without substrate cooling in a nitrogen-rich atmosphere after deposition; (b) sputtering power between 180 W and 300 W, nitrogen-argon ratio of 16:4, and with substrate cooling in a nitrogen-rich atmosphere after deposition. The refractive index was obtained with an ellipsometer (*alpha-SE Ellipsometer, J.A. Woollam Co.*), applying the *Tauc-Lorentz* model.

with a nitrogen-argon ratio of 13:7, and a pressure of 6×10^{-3} mbar during deposition. A deposition rate ranging from 0.4 Å/s to 1 Å/s was measured.

Secondly, as an attempt to improve the thin-films refractive index, it was performed the deposition of SiN thin-films on silicon substrates with power supplies ranging from 180 W to 300 W, with a nitrogen-argon ratio of 16:4, a pressure of 6×10^{-3} mbar during deposition, and applying the substrate cooling in a nitrogen-rich atmosphere after sputtering, which consisted of a constant flow rate of 16 sccm of nitrogen until the samples reached 25 °C. A deposition rate ranging from 0.7 Å/s to 0.9 Å/s was measured. Table 1 summarizes all the performed SiN thin-film depositions.

Ellipsometry of the SiN thin-films deposited in silicon slices was performed using a commercial ellipsometer (*alpha-SE Ellipsometer, J.A. Woollam Co.*) and applying the *Tauc-Lorentz* model. The general form of the model is given by (1):

Table 2
Fitting parameters from the *alpha-SE Ellipsometer* software of the deposited SiN thin-films.

Power (W)	Fitting thickness (nm)	Fitting roughness (nm)	MSE	Substrate cooling in a nitrogen-rich atmosphere
100	350.93 ± 0.208	3.76 ± 0.375	10.903	No
150	408.80 ± 0.271	6.37 ± 0.461	14.896	
200	208.46 ± 0.192	0.00 ± 0.518	9.697	
180	243.18 ± 0.534	2.49 ± 0.795	10.331	Yes
200	236.27 ± 0.583	5.41 ± 0.791	18.133	
230	223.37 ± 0.229	1.97 ± 0.245	9.893	
250	223.69 ± 0.192	0.84 ± 0.198	8.305	
300	219.82 ± 0.181	1.48 ± 0.184	8.030	

$$\varepsilon_{2TL}(E) = \begin{cases} \dfrac{AE_0C(E-E_g)^2}{(E^2-E_0^2)^2+C^2E^2} \bullet \dfrac{1}{E}, E > E_g \\ 0, E \leq E_g \end{cases} \tag{1}$$

where ε_2 is the imaginary part of the dielectric function, the subscript *TL* indicates that the model is based on the Tauc joint density of states and the Lorentz oscillator, E is the photon energy, A is the transition amplitude, C is the broadening term, E_0 is the peak transition energy, and E_g is the optical band gap. The four fitting parameters A, C, E_0, and E_g are all in units of energy (eV) [23]. In the software of the ellipsometer, the *Tauc-Lorentz* model was used to fit these parameters, extracting the SiN refractive index and other thin-film features (thickness and roughness).

SiN thin-films were also analyzed by EDS and AFM using commercial equipment, *EDAX - Pegasus X4M (EDS/EBSD)* at *SEMAT – University of Minho* and *Dimension Icon – BRUKER* at *3B's Research Group – University of Minho*, respectively.

3. Results and discussion

Fig. 1 shows the refractive index curves for all the deposited SiN thin-films, as a function of the wavelength, for different sputtering powers, different nitrogen-argon ratios, and samples without and with substrate cooling in a nitrogen-rich atmosphere after deposition. Table 2 shows the other fitting parameters of all the samples, thickness and roughness, with the respective mean square error (MSE) of the fitting process. As can be seen in Fig. 1, the increase in the sputtering power leads to the increase of the SiN refractive index, as expected according to the literature [20]. However, this increase is more significant in Fig. 1 (b) compared with Fig. 1 (a), presumably because there is a higher ratio of nitrogen-argon during deposition and also the effect of the substrate cooling in a nitrogen-rich atmosphere after deposition. These changes possibly led to thin-films with higher packing density, which in turn led to a higher resistance to oxidation after deposition [8]. Contrarily, the lower refractive indices present in Fig. 1 (a) can be explained by lower packing density thin-films with stronger tendency of the silicon to bond with oxygen than with nitrogen. Some silicon atoms reacted with the oxygen, potentially originating from the inner wall of the deposition chamber, residual gas, or gas leaks during deposition [13]. Thus, at 400 nm the maximum SiN refractive index obtained was approximately 1.906 considering a sputtering power of 300 W, a nitrogen-argon ratio of 16:4, and a substrate colling in a nitrogen-rich atmosphere after deposition. This result is close to the refractive index of stoichiometric Si_3N_4

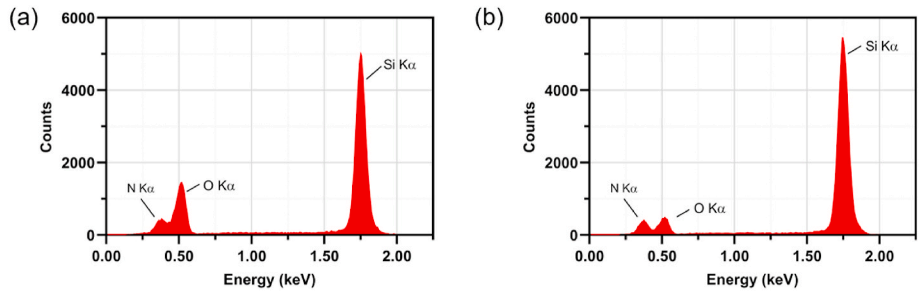

Fig. 2. EDS spectrum of SiN samples: (a) deposited at 200 W and with a nitrogen-argon ratio of 13:7, and without substrate cooling in a nitrogen-rich atmosphere; and (b) deposited at 300 W and with a nitrogen-argon ration of 16:4, and with substrate cooling in a nitrogen-rich atmosphere.

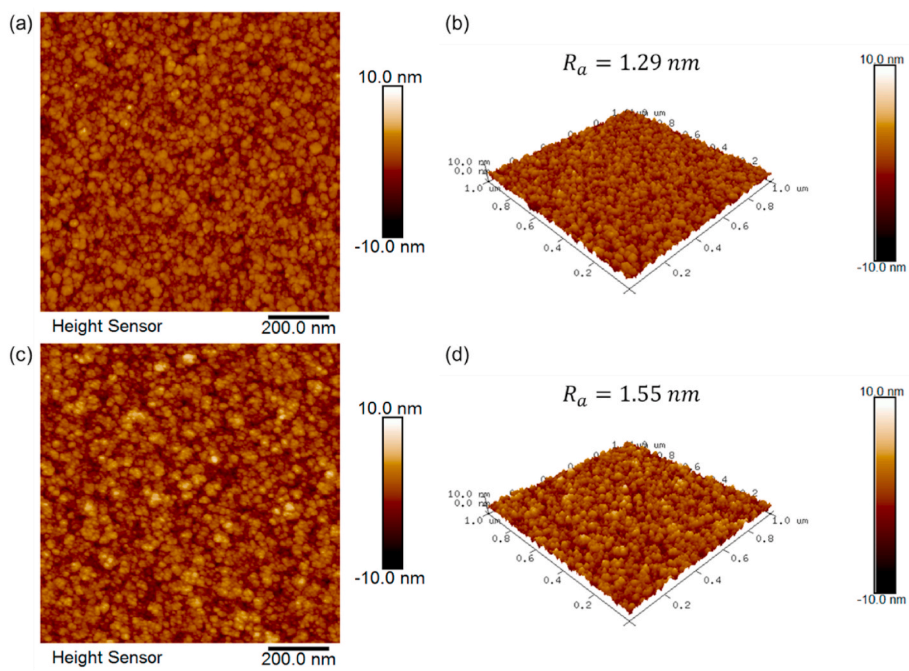

Fig. 3. AFM topography of SiN samples: (a) 2D topography and (b) 3D topography of the same sample as Fig. 2 (a); (c) 2D topography and (d) 3D topography of the same sample as Fig. 2 (b).

of 2.073 at 400 nm [13]. Moreover, literature results about the optimization of SiN refractive index deposited by RF reactive sputtering stated a refractive index ranging from approximately 1.6 to 2.7 at ≈ 400 nm, depending on the films deposition parameters [20,22]. It is also reported in the literature that in PE-CVD SiN_x films, with an H content of less than 5 %, the refractive index ranges from 1.859 to 2.01 at 630 nm, dependent on increasing substrate temperature, plasma power, and precursor flow and from 2.15 to 2.31 at 632 nm, dependent on increasing film thickness [3].

Fig. 2 shows the EDS spectra of SiN thin-films (performed at 10 kV): (a) deposition at 200 W with a nitrogen-argon ratio of 13:7 and without substrate cooling in a nitrogen-rich atmosphere after deposition; and (b) deposition at 300 W with a nitrogen-argon ratio of 16:4 and with substrate cooling in a nitrogen-rich atmosphere after deposition. In the sample of Fig. 2 (a), EDS analysis revealed silicon (Si) with an atomic percent of 39.86 %, nitrogen (N) of 25.79 %, and oxygen (O) of 34.35 %. In the sample of Fig. 2 (b), EDS analysis revealed Si with an atomic percent of 53.77 %, N of 30.47 %, and O of 15.76 %. Thus, it can be concluded that a higher sputtering power and a higher nitrogen-argon ratio during deposition, and the substrate cooling in a nitrogen-rich atmosphere after deposition reduced the thin-film oxidation, which is in accordance with the optical characterization of the SiN thin-films (higher refractive indices).

The surface roughness of the sputtered films was also evaluated by AFM in tapping mode as a direct measurement. As can be seen in Fig. 3, an area of 1 × 1 μm^2 was analyzed. Fig. 3 (a) and Fig. 3 (b) were obtained by measuring the same sample as Fig. 2 (a), in which an average surface roughness (R_a) of 1.29 nm was acquired. Fig. 3 (c) and Fig. 3 (d) were obtained by measuring the same sample as Fig. 2 (b), in which an R_a of 1.55 nm was acquired. The increase of 0.26 nm in R_a can be explained by the larger clusters of SiN present in the sample of Fig. 3 (c) and Fig. 3 (d).

4. Conclusions

Along this paper, several SiN thin-films were deposited by RF reactive sputtering under different sputtering powers (180 W–300 W), with a high nitrogen-argon ratio (16:4), and performing substrate cooling in a nitrogen-rich atmosphere immediately after deposition. The SiN thin-films were assessed through ellipsometry to determine their refractive indices, through EDS to determine their element composition, and through AFM to acquire their surface topography. The EDS and AFM analysis led to the conclusion that the SiN thin-films deposited with higher sputtering power (300 W), higher nitrogen-argon ratio (16:4), and on which a substrate cooling in a nitrogen-rich atmosphere was applied had an increased atomic percent of nitrogen, a decreased atomic percent of oxygen, and an R_a of 1.55 nm. The maximum obtained SiN refractive index at the conditions referenced above was 1.906 at 400 nm. This is the first study reporting the substrate cooling in a nitrogen-rich atmosphere after reactive sputtering of SiN thin-films.

CRediT authorship contribution statement

João R. Freitas: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **Sara Pimenta:** Writing – review & editing, Writing – original draft, Validation, Supervision, Investigation, Formal analysis, Conceptualization. **Vítor H. Rodrigues:** Investigation, Conceptualization. **Manuel F. Silva:** Supervision, Investigation, Conceptualization. **José H. Correia:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgments

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