Measurement of silicon suboxide films thickness synthesized by the gas-jet electron beam plasma chemical vapor deposition method

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Abstract. The amorphous silicon suboxide thin films were synthesized by the gas-jet electron beam plasma chemical vapor deposition method. The thickness of the thin films was obtained using a cross-section SEM image (destructive) and an analysis of interference effects in the IR transmission spectra (non-destructive). The film thicknesses obtained by the cross-section SEM images were about 600 nm for all samples. An approximation of the silicon suboxide film thickness was made using a Gaussian distribution, which showed good agreement with the experimental values. The thickness obtained from the analysis of the IR transmission spectra increases from 400 nm to 500 nm with increasing Gmix. It was found that the oxygen concentration of the thin films decreases from 45 to 22% with an increase in Gmix.

1. Introduction
Silicon suboxide thin films (SiOx, 0 < x < 2) are widely used as a raw material in the fabrication of thin-film transistors in microelectronics [1], as a passivation [2] and buffer layer [3] in solar cells, as the initial silicon-containing material to obtain nano- [4] and polycrystalline silicon [5], and etc. Therefore, accurate determination of thickness and chemical composition in these films is essential.

The methods for measuring the film thickness can be divided into two groups: non-destructive and destructive ones. The non-destructive methods include Fourier-transform infrared spectroscopy (FTIR) [6], ellipsometric measurements [7], UV–Visible spectroscopy measurements of transmittance spectra [8] and Rutherford backscattering spectrometry measurements [9]. The most widely used destructive methods are the scanning electron microscopy (SEM) [10, 11] and transmission electron spectroscopy (TEM) [12]. The method that can be both destructive and non-destructive is the measurement of film thickness using a profilometer [13, 14].

In this paper amorphous hydrogenated SiOx (a-SiOx:H) thin films were synthesized by the gas-jet electron beam plasma chemical vapor deposition (GJ EBP CVD) method. The cross-section SEM image (destructive) and analysis of interference effects in the IR transmission spectra (non-destructive) were used to estimate the thicknesses of the synthesized thin films. For the first time, the comparison of thicknesses obtained by these methods was made for a-SiOx thin films.

2. Experimental details
In this work a-SiOx:H thin films with the thicknesses of about 600 nm were prepared by GJ EBP CVD method onto clean monocrystalline silicon (c-Si) substrates at the synthesis temperature of 260°C. A detailed description of this method is given in [15]. Different stoichiometry characteristics of the films were obtained by varying the flow rates of 5%SiH4+95%Ar mixture (Gmix) at the constant oxygen flow
rate of 5 sccm into the vacuum chamber. The flow rates $G_{\text{mix}}$ were 20, 60, 80, 100 sccm and the flow rate of hydrogen was 600 sccm. Synthesis time was 40 min for each sample. The pressure in the vacuum chamber was 20 Pa during synthesis. The gas mixture was activated by an electron beam with energy of 1000 eV and a current of 70 mA.

Cross-section SEM images were used to measure the thickness of the films. SEM images were obtained by using a JEOL JSM-6700 F microscope. The samples were pre-cooled in liquid nitrogen to obtain a high quality cross-section of cleavage. FTIR spectra were used to measure non-destructively the thickness and composition (oxygen concentration) of the films [16]. FTIR spectra were recorded using a Bruker IFS-113V Fourier spectrometer in the wavenumber range from 400 to 4000 cm$^{-1}$. Because of the multiple reflections from the “air/thin film” and “thin films/c-Si” interfaces interferential picture was obtained in FTIR spectra. It is possible to obtain thickness and refractive index of the films by interferential bands analysis [6]. The obtained thin films have a gradient of oxygen concentration from the center to the periphery, and due to the deposition conditions, these films also have a thickness gradient with a normal thickness distribution.

3. Results and discussion

Figure 1 shows the cross-section SEM images of samples synthesized with $G_{\text{mix}}$ values of 20, 60, and 100 sccm. The figure shows that the a-SiO$_x$:H film has internal structure, which we associate with the pores in the material. Moreover, the porosity of the material increases with an increase in $G_{\text{mix}}$.

![Figure 1](image)

The measurements of the thickness were carried out every 3 mm along the sample. Since the sample size was 18 mm, five values of thickness in the different points were obtained for each sample. The experimentally obtained thicknesses for $G_{\text{mix}} = 60$ sccm are presented in Figure 2. As it can be seen, the maximum thickness is reached at the center and decreases at the periphery. As it is known, the synthesis parameters affect the distribution of film thickness along the sample.

The main parameters are flow rate and type of gas, as they influence the gas dynamics of the supersonic jet [17]. The gas dynamics of the jet determines the interaction region of the jet with the surface and affects the distribution of the active components on the sample surface. The next parameters are the energy and the focusing of the electron beam. These parameters form the activation zone of the supersonic jet. On the one hand, the higher the electron energy, the more molecules and atoms can be excited by the primary electrons. But, on the other hand, an increase in the electrons energy leads to a decrease in the reaction cross section with molecules of the gas. The maximum cross-section of the reaction for monosilane (SiH$_4$) molecules is around 100 eV. In addition, the article [18] showed that the interaction of argon with electrons leads to the formation of argon atoms in metastable state and an increase in the amount of fast secondary electrons, which leads to the rise of the number of activated molecules from which the film grows. It was concluded that the distribution of silicon suboxide film thickness is formed as a result of the influence of interdependent synthesis
parameters, so the thickness distribution on the sample can be described using the Gaussian distribution.

An approximation of the silicon suboxide film thickness was made using a Gaussian distribution, which is shown in Figure 2. This procedure was done for all the samples. A description of thickness using Gaussian distribution showed good agreement with the experimental values and the peak width at half maximum (FWHM) was 20 ± 0.3 for all films. Several measurements of thickness were made for one point, and the resulting error was about 2%.

Since the thickness measurement by the cross-section SEM image is a destructive method, the thickness measurement was taken by the common procedure of analysis of interference effects in the transparent region of the IR transmission spectra. A typical comparison between an experimental spectrum and a simulated transmittance baseline is shown in Fig. 3 for sample with $G_{\text{mix}} = 60$ sccm. Simulated transmittance baseline was obtained using the software described in the article [19]. For sample with $G_{\text{mix}} = 60$ sccm, the good agreement between the calculated and experimental curves was obtained for a thickness of 400 nm. The thickness of silicon suboxide thin films for all samples, obtained using the cross-section SEM method and procedure of analysis of interference effects in the IR transmission spectra are shown on Figure 4.

The thickness of the thin films was obtained by cross-section SEM image and by analysis of interference effects in the IR transmission spectra, as a function of $G_{\text{mix}}$.

The oxygen concentration in a-SiO$_x$ thin films on $G_{\text{mix}}$. 

![Figure 2](image1.png)  
**Figure 2.** The thickness of the thin films was obtained by SEM image cross-section for the sample with $G_{\text{mix}}=60$ sccm and approximation by Gaussian distribution.

![Figure 3](image2.png)  
**Figure 3.** Optical density and calculated interference spectrum for sample with $G_{\text{mix}}=60$ sccm.

![Figure 4](image3.png)  
**Figure 4.** The thickness of the thin films was obtained by cross-section SEM image and by analysis of interference effects in the IR transmission spectra, as a function of $G_{\text{mix}}$.

![Figure 5](image4.png)  
**Figure 5.** The oxygen concentration in a-SiO$_x$ thin films on $G_{\text{mix}}$. 

It can be seen that the average thickness obtained from the cross-section SEM image is 600 nm and varies slightly depending on $G_{\text{mix}}$. While the thickness obtained from the analysis of the IR transmission spectra increases from 400 nm to 500 nm with increasing $G_{\text{mix}}$. Probably, difference in the thickness values of SEM and FTIR is due to the low accuracy of FTIR measurements. Firstly, the region of the FTIR measurements is 5 mm in diameter and the film thickness changes by about 50 nm (Fig. 2) in this region. Secondly, the results are influenced by the optical heterogeneity of the structure (silicon-rich clusters, oxygen-rich clusters, voids in the structure as shown in the article [16]), while the formulas for the film thickness calculating assume the use of optically uniform structures.

Usually, the FTIR method is used to determine the concentration of oxygen and hydrogen for a-SiO$_x$:H films. The hydrogen was not detected in the FTIR spectra for the sample with $G_{\text{mix}} = 20$ sccm. For the other samples the hydrogen concentration was about 2%. As shown in Figure 5, the oxygen concentration decreases from 45 to 22% when the $G_{\text{mix}}$ increases. A possible explanation for this effect is an increase in the density of the gas jet because of an increase in the flow rate of such a heavy gas as argon. Accordingly, the increase in density leads to a more difficult diffusion of oxygen molecules from the background gas into the growth area of the film.

**Conclusion**
The amorphous silicon suboxide thin films were synthesized by the gas-jet electron beam plasma chemical vapor deposition method. The film thicknesses obtained by the cross-section SEM images were about 600 nm for all samples. An approximation of the silicon suboxide film thickness was made using a Gaussian distribution, which showed good agreement with the experimental values. The thickness obtained from the analysis of the IR transmission spectra increases from 400 nm to 500 nm with increasing $G_{\text{mix}}$. It was found that the oxygen concentration of the thin films decreases from 45 to 22% with an increase in $G_{\text{mix}}$.

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