Formation of thin silicon films on soda-lime silica glass surface by magnetron sputtering deposition

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Magnetron high-power impulse sputtering was used for deposition of H_2-free amorphous thin Si films in Ar atmosphere on a flat surface of soda-lime silicate glass at different values of the substrate temperature and bias potential to glass. The surface morphology of the deposited films and their transverse structure were studied by scanning electron and atomic-force microscopy. For various deposition regimes, the appearance on the film surface spherical craters with diameters of several hundred nanometers and Si nanowires towering over a film were observed. The degree of transparency of the films in the visible spectral range was also analyzed. This work demonstrates that magnetron sputtering offers a good potential for depositing the transparent films for silicon-based photonics.

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1. Introduction

The search for a solution to the energy problem for efficient conversion of solar radiation into electricity is one of the priority areas of both fundamental and applied research. At present, in practice, to ensure the required high level of solar radiation absorption, devices made of sufficiently thick crystalline c-Si plates with thickness from 180 to 300 μm are used. In this case, the main costs for the manufacture of solar cells are determined by the growing and subsequent technological processing of thick-layer c-Si substrates. Therefore, it seems relevant and economically important to create optically transparent solar cells with thin (<0.5 μm) Si films (TSiFs) of an amorphous or polycrystalline structure, which could also be used in multilayer photoconverters [1, 2]. For thin-layer solar cells, the area of their functional application is expanding when they are integrated with modern compact ultra-lightweight optoelectronic microcircuits and Si-based devices for use in the aero- and space industry. The characteristics of such devices largely depend on the structural features and optical absorption properties of TSiFs [2].

Deposition techniques including ion bombardment or not, such as in rf plasma-enhanced chemical vapor deposition, hot wire chemical vapor deposition were used to prepare TSiF photovoltaic materials with different microstructures. However, the magnetron sputtering proves for the deposition as another choice [2], and very little is reported on the microstructure of the films obtained by this technique. One of the earliest works on the formation of TSiF by the magnetron sputtering deposition, apparently, was published in 1985 [3]. When fabricating TSiF, various experimental parameters were used for this method, such as the substrate temperature (T_{sub}), the type of gas sputtering atmosphere, bias potential (U_{bp}), power densities at the target, deposition rate (nm/min) etc. For example, in the work [3] amorphous a-Si:H films were deposited in Ar-H_2 atmosphere with a H_2 content of 3 and 20% at a substrate temperature between 250 and 300 ºC. For various applications in the TSiF magnetron sputtering *process,* various types of specific target material substrates were used: monocrystalline c-Si [2, 3], suprasil-grade quartz [3], bismuth-coated ITO glass [4], Ni prelayers on SiO_2 [5] or Ag films [6] and so on.

This work is devoted to the study of the surface morphology and optical properties of hydrogen-free TSiFs formed by magnetron sputtering deposition on the surface of optically transparent soda-lime silica glass at various technological parameters such as T_{sub} and U_{bp}. Note that if in the scientific literature enough attention was paid to the obvious parameter T_{sub}, which determines the property TSiFs, then the importance of the role for U_{bp} began to be closely considered relatively recently [7]. Thus, in the present work, various technological parameters of magnetron sputtering deposition were used for the preparation of TSiFs, and a comparative analysis of the obtained film samples was carried out.

2. Experimental

As substrates for the deposition of TSiFs, flat polished plates of soda-lime silicate glass with a size of 24 × 24 mm^2 and a thickness of 0.18 mm were used. To remove residual organic contaminants from the surface of the substrates and increase the adhesion of deposited TSiFs, preliminary ionic cleaning of glasses was carried out for 10 min using an ion source "Radical" [8]. Next operating parameters for cleaning were selected: Ar gas pressure $4.5 \cdot 10^{-4}$ Torr, discharge current and voltage I = 20 mA and U = 2.4 V respectively. Cleaning was carried out in a vacuum chamber of a UVN-2M magnetron sputtering setup, in which TSiFs were also later deposited in an Ar atmosphere at a pressure of $5.2 \cdot 10^{-4}$ Torr. To maintain a constant gas pressure during the Ar plasma generation during fabrication of TSiFs, an original feedback system

was applied, described in detail in the work [7]. An undoped *c*-Si wafer with a diameter of 10 cm was selected as a target for sputtering.

A high-power impulse magnetron sputtering approach with a frequency of 50 kHz, a duty cycle of 80 % at a constant value of I = 610 mA, and U = 710-720 V was used for the formation of TSiFs. The duration of TSiF deposition was 2.5 min. The discharge power was kept constant and set in the range of 400-600 W. The distance from the target to the substrate surface was 80 mm. During the deposition of TSiFs, the specified parameters T_{sub} and U_{bp} were chosen and the corresponding values of which for each of the samples are given in Table 1.

The surface morphology of TSiFs were studied with atomic-force microscope (AFM) Dimension FastScan (Bruker) by quantitative nanomechanical mapping and with scanning electron microscope (SEM) Merlin (Carl Zeiss) using an In-Lens secondary electron detector at an accelerating voltage of 5 keV and current density 300 pA. For similar SEM conditions, but tilting the samples relative to the normal electron beam by an angle of 70° an observation of transverse cleavages of the samples as well as their structural measurements by electron backscattered diffraction detector HKL NordLys detector (Oxford Instruments) were carried out. The optical transmission spectra of the samples were measured in the range 350-1100 nm with a waveguide spectrometer AvaSpec-2048 (Avantes).

Table 1. Sample numbering and corresponding T_{sub} and U_{bp} values used during magnetron sputtering deposition of TSiF

Sample №	T_{sub} (ºC)	U_{bp} (V)
1	Room	0
2	Room	-200
3	380	0
4	480	-175
5	250	-80

3. Results and discussion

Figs. 1 and 2 reveal the SEM and AFM images of TSiF surfaces. AFM-images are helpful to observe morphology features with high topography details on small surface sample area. As follows from the SEM-images of sample cross-cleavages in Fig. 1 (b, d, f, h, g), the thicknesses of deposited TSiFs varied in the range of 100-200 nm. As an example, the inset in Fig 1 (b) shows an enlarged cross-sectional view of the TSiF film, allowing direct measurement of the thickness. The observation of the electron backscattered diffraction patterns indicates that all TSiFs formed at different values of T_{sub} and U_{bp} are characterized by an amorphous *a*-Si structure.

From SEM- and AFM-images of Sample 1 (Fig. 1 a, b; Fig. 2 a, b) it follows that as a result of magnetron sputtering deposition at room temperature and the absence of U_{bp} (Table 1), TSiF is formed with a smooth surface, on which small spherical craters with a diameter of about 200–400 nm are observed. The presence of such large volume defects as craters in the TSiF surface region reduces the practical attractiveness of this film for solar cells, despite the presence of rather large smooth surface areas on the sample 1. However, after deposition without heating but with U_{bp} = -200 V, fabricated TSiF is (Sample 2) also with a smooth surface area, which is much freer from defects and irregularities (Fig. 1 c, d; Fig 2 c, d) in contrast to sample 1. Moreover, the craters formed in this case on the surface of the Sample 2 are characterized by noticeably smaller diameters and depths compared to Sample 1. Additionally, on the surface of Sample 1 there are some raised small irregularities, which, apparently, are artifacts (dust grains) that fell on the surface during the preparation of sample for SEM observations.

In the event that magnetron deposition occurs on a heated substrate up to T_{sub} = 380 ºC in the absence of U_{bp} (Sample 3), then, as seen from the microscopic data (Fig. 1 e, f; Fig. 2 e, f), the sample surface is very rough with the presence of large irregular Si formations. However, the spherical craters observed for Samples 1 and 2 are absent for Sample 3. If the substrate is heated only to T_{sub} = 480 ºC and U_{bp} = -175 V (Sample 4), then again, as in the case of Samples 1 and 2, the formation of many craters is observed on the surface of sample 4 (Fig. 1 g, h; Fig. 2 g, h) with a noticeably higher concentration, covering almost half from the free smooth surface of the sample. The diameters of the spherical craters are also quite large, ranging in size from 200 to 500 nm.

Of particular interest is Sample 5, which fabricated at the average of the values used in the present experiments: T_{sub} = 250 ºC and U_{bp} = -80 V. On the TSiF surface (Fig. 1 i, g; Fig. 2 i, g) the presence of a multitude of Si nanowires as 200-250 nm long and about 10-20 nm in diameter were observed, which early not presented in the scientific literature on magnetron sputtering deposition of TSiFs. These nanowires evenly cover the entire area of Sample 5 and with orientation in one direction.

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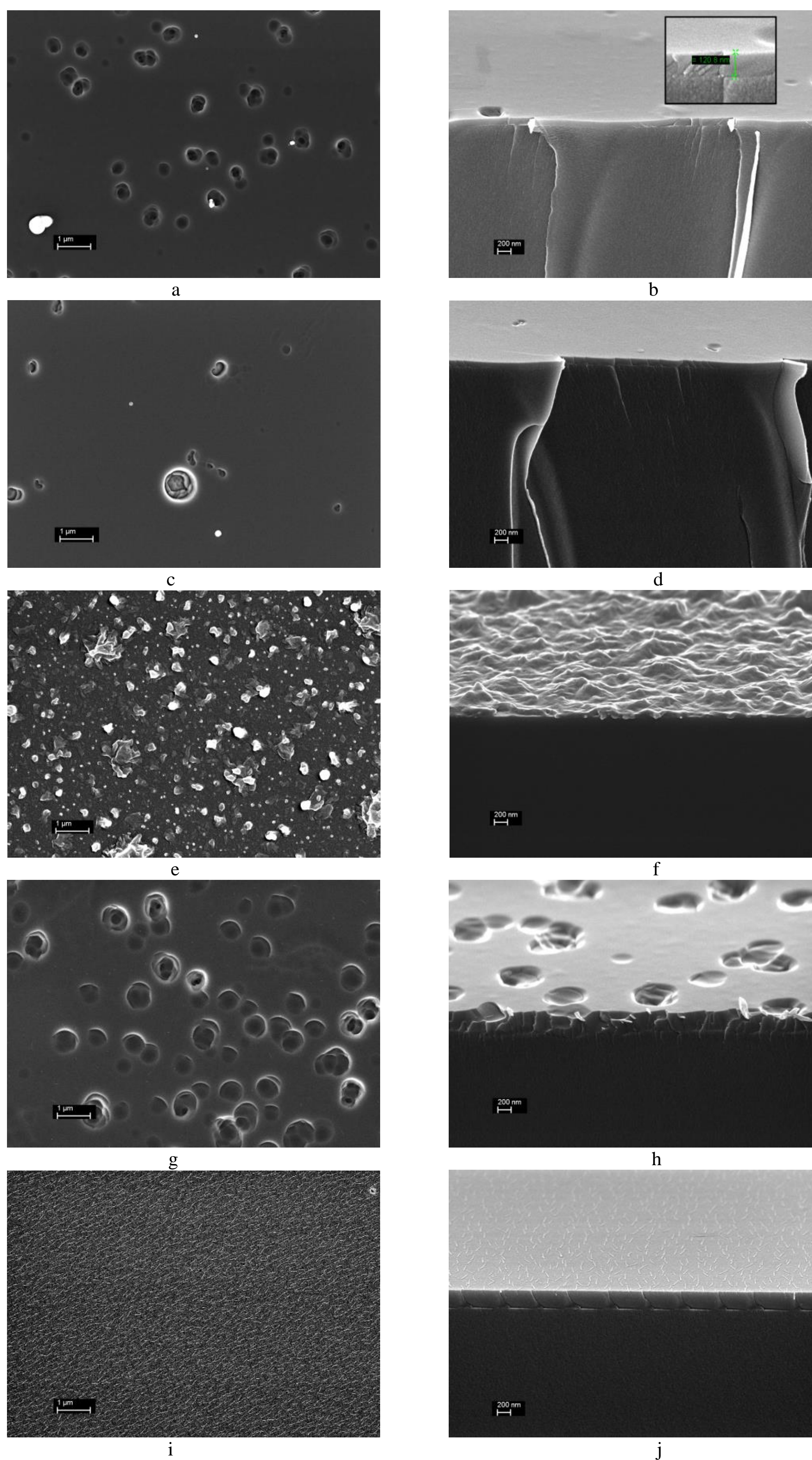

Fig. 1. SEM-images of a-Si films formed by magnetron sputtering deposition for various experimental parameters: (a, b) Sample 1, (c, d) Sample 2, (e, f) Sample 3, (g, h) Sample 4 and (i, j) Sample

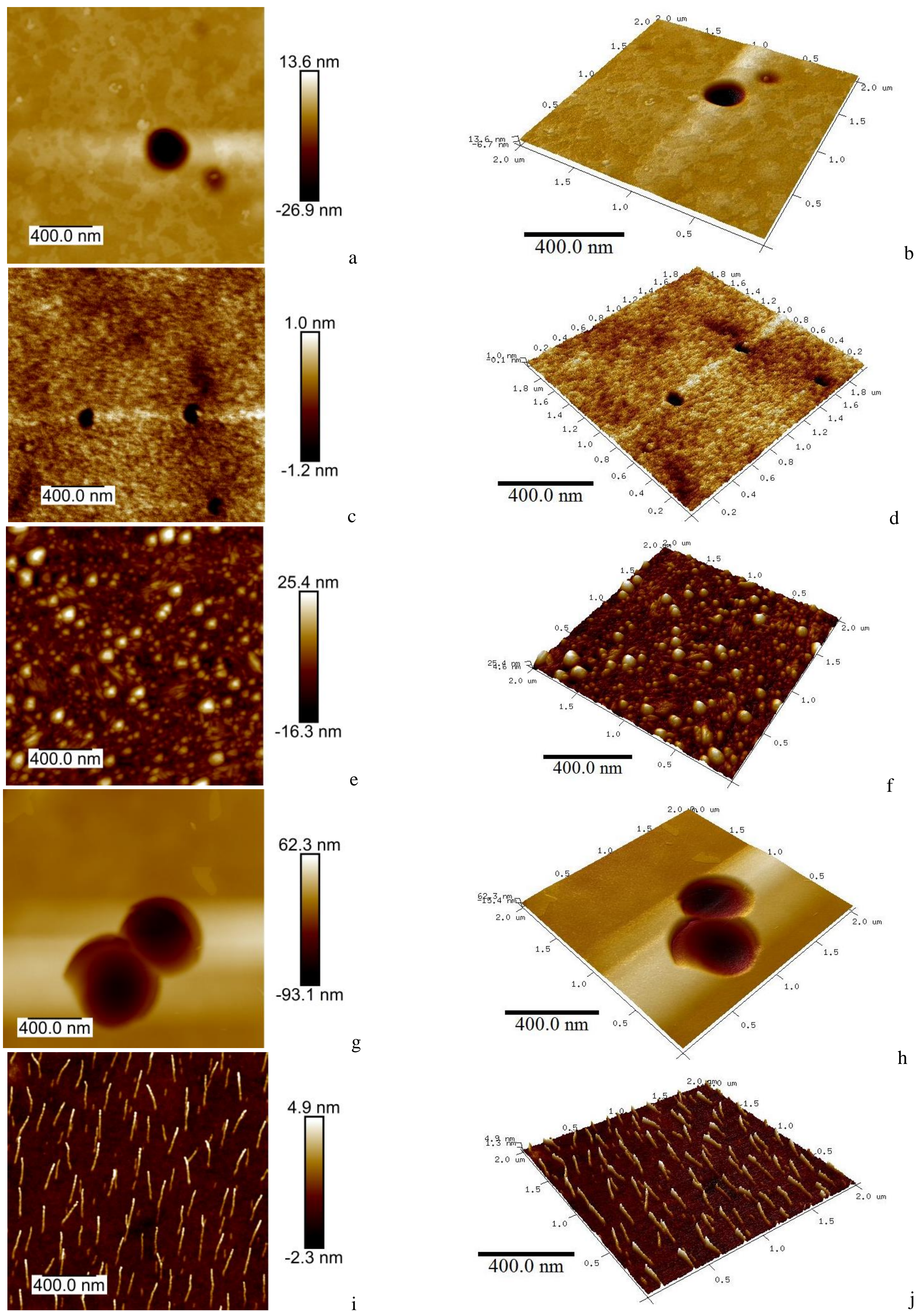

Fig. 2. AFM-images of a-Si films formed by magnetron sputtering deposition for various experimental parameters: (a, b) Sample 1, (c, d) Sample 2, (e, f) Sample 3, (g, h) Sample 4 and (i, j) Sample 5 (color online)

Of particular interest is Sample 5, which fabricated at the average of the values used in the present experiments: T_{sub} = 250 ºC and U_{bp} = -80 V. On the TSiF surface (Fig. 1 i, g; Fig. 2 i, g) the presence of a multitude of Si nanowires as 200-250 nm long and about 10-20 nm in diameter were observed, which early not presented in the scientific literature on magnetron sputtering deposition of TSiFs. These nanowires evenly cover the entire area of Sample 5 and with orientation in one direction.

The mechanisms of the formation of such large defects in the form of spherical craters and raised by nanowires on the surface of the TSiF obtained by magnetron sputtering deposition were not previously described and therefore require a deeper study. However, evaluating the morphology and quality of TSiF surfaces depending on the used technological parameters, it possible to conclude that for the practical use of TSiFs for the design of solar cells and photodetector devices, Sample 2 (T_{sub} = 250 ºC and U_{bp} = -80 V) is the most attractive, because this TSiF is characterized by a large smooth surface area with a minimum number of defects.

To assess the ability to use the obtained TSiFs as optically transparent elements, their optical transmittance spectra were measured and presented in Fig. 3. As seen from the above spectra, all the obtained TSiFs are demonstrated a certain degree transparency in the visible optical range, in which the bulk Si material almost completely absorbs light. Observed in Fig. 3 curves represent spectral dependences of alternating maxima and minima depending on a particular sample. This typical spectral character is determined by the interference of light from TSiF located on the surface of the transparent substrate [9]. The difference in the numbers of maxima and minima could be explained by the variations of TSiF density, the corresponding optical absorption and refraction constants, the presence of large surface defects and the Rayleigh scattering of light caused by them [10], as well as the discrepancy between the TSiF thicknesses, as seen from measurements in sample cleavages (Fig. 1). Selected Sample 2 with best surface morphology is characterized with very wide transparence band with maximum near 800 nm.

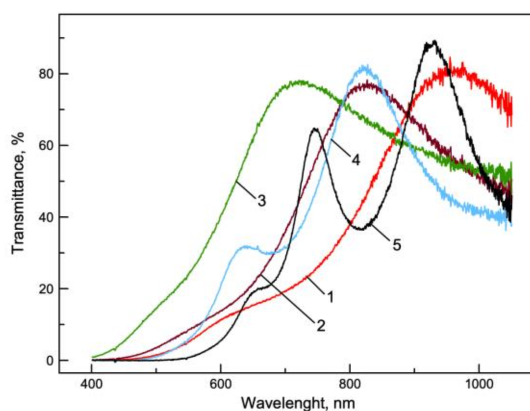

Fig. 3. Optical transmission spectra a-Si films formed by magnetron sputtering deposition for various experimental parameters: (1) Sample 1, (2) Sample 2, (3) Sample 3, (4) Sample 4 and (5) Sample 5 (color online)

4. Conclusion

Thus, in present study magnetron high-power impulse sputtering was used for deposition of TSiF on the surface of soda-lime silicate glass substrate. By varying the technological parameters T_{sub} and U_{bp}, TSiFs with different morphology and a set of surface defects were obtained. It was found that the most high-quality and low-defect TSiF qualitatively suited for practical applications was fabricated at T_{sub} = 250 ºC and U_{bp} = -80 V. Also, it is shown that all films have a sufficiently high optical transparency in the visible range of the spectrum.

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