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Magnetron sputtered silicon thin films for solar cell applications

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Abstract

One of the important directions of research in photovoltaics is the development of new thin-film technology, which can replace the currently used, more expensive bulk silicon technology. The article discusses the findings from research focused on optimizing the parameters for the deposition of silicon thin films with P-type electrical conductivity for applications in photovoltaics. The growth rate was determined depending on the change in substrate temperature using reflectometry and the influence of deposition time on optical properties was determined using UV/VIS spectroscopy. Photovoltaic structures were made on substrates with an ITO layer and their electrical parameters were measured. The authors applied the magnetron sputtering method to deposit the layers, selecting it over the commercially used the chemical vapor deposition (CVD) method. This replacement could alleviate the necessity for high temperatures and broaden the potential applications of thin-film solar cells.

1. Introduction

Obtaining electricity from non-renewable energy sources has a negative impact on the environment and is becoming more and more expensive. Obtaining energy from renewable sources is limited by scientific and technical development and costs, but reports from the energy industry indicate that within a decade the costs of photovoltaic solutions will approach conventional solutions (Fig. 1) [1–3].

Therefore, there is a real need to conduct research on increasing the performance and minimizing the production costs of solar cells [3–5]. Currently, the solar market is dominated by silicon solar cells. Figure 2 shows the share of individual solar technologies in the photovoltaic market. The chart was prepared based on data from the "REGlobal" and "GlobalData" portals as of Feb. 2nd, 2024.

Silicon is the second most common chemical element on Earth and its properties are well known due to its important role in the electronics industry. The expeditious expansion of the photovoltaic industry based on crystalline silicon photovoltaic cells has resulted in the development of new technologies that increase their production while reducing the consumption of this element. Solar cells made from monocrystalline silicon achieve an efficiency of 21.4%

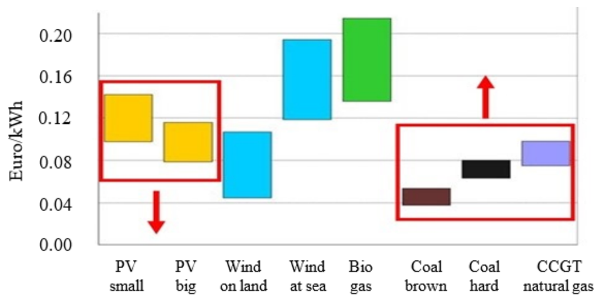

Fig. 1. Changes in energy costs from individual sources (own work based on data from the "Our World in Data" and "European Central Bank" portals).

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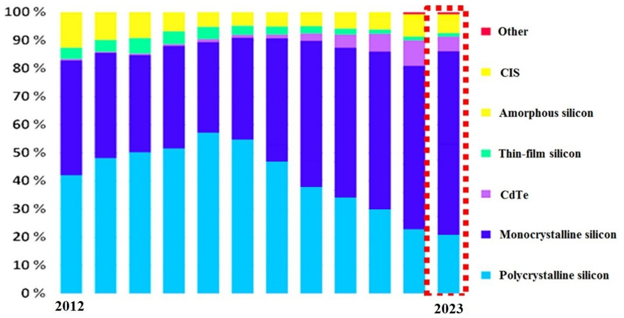

Fig. 2. Percentage market share of individual photovoltaic technologies (own study based on data from the "REGlobal" and "GlobalData" portals).

in series production and show stable operating conditions in long-term exploitation [4–7]. In May 2024, an efficiency record for commercial monocrystalline solar cells of 27.3% (Longi) was achieved [8]. However, the development of new technologies utilizing solar cells requires further research. A key factor that can significantly accelerate progress in this field of science is expanding the scope of solar cell applications while simultaneously reducing production costs. Although silicon is a widely available material and the cost of production is reduced by lowering the thickness of silicon wafers, this state has its limitations. Therefore, high hopes are placed on thin-film technology, which enables the reduction of material consumption, the use of a low-cost substrate, and the extension of the use of photovoltaics by reducing the weight of the finished product and flexibility. Hence, thin-film photovoltaic cells have become a viable substitute for conventional bulk silicon solar cells. Their advantage is the lower direct cost of semiconductor materials. Additionally, an important advantage is the possibility of producing them on various substrates, including flexible ones [8–10].

The current and most promising solutions in the thin-film technology are based on amorphous silicon, which allows to obtain a solar cell with an efficiency of about 12% using a multi-junction [9, 10]. Another solution within the thin-film technology is using hydrogenated amorphous silicon (a-Si:H) solar cells. Hydrogen is introduced into the amorphous silicon structure to passivate dangling silicon bonds, reducing the number of defects in the material and improving its electrical properties and mechanical characteristics, such as flexibility. In this solution, 14% efficiency was achieved [9, 10]. Typically, the chemical vapor deposition (CVD) method and its variations are applied, which requires high temperatures and hazardous gases. In addition, this solution limits the range of substrates used [9–11].

Magnetron sputtering (MS) is one of the physical vapor deposition (PVD) techniques. The target material is sputtered with ions, usually argon. During this process, a source-target material is dislodged due to either a direct current (DC) glow discharge or a flow of radio frequency (RF) current assisted by a plasma. Moreover, a magnetic field is employed to enhance the ionization process, which increases the number of ions bombarding the source target. This adjustment in the process facilitates the accelerated thin-film formation on the substrate surface. Notably, this method provides exact planning of the thickness of the produced thin film, even at the nanometer scale, resulting in the deposition of high-quality layers with a uniform material structure and low roughness [10–12]. The MS technique has recently gained increasing attention compared to CVD-based methods due to its inherent advantages. These include simplicity, low-temperature processing, higher silicon use, and avoidance of toxic materials such as SiH_4. These benefits make MS a cost-effective and environmentally friendly fabrication method, providing high process uniformity and scalability [12–16].

The plasma-assisted MS method could be used to reduce high temperature limitations. The article presents the research results on the possibility of depositing silicon thin films with a P-type conductivity. Those layers deposited by MS can be used in the thin-film solar cell technology. The authors used the MS method instead of the commercially used CVD method to deposit the layers. This may eliminate the problem of the need to use high temperatures and expand application possibilities of thin-film solar cells. Additionally, numerous companies are involved in producing materials for PVD and companies using PVD techniques. Therefore, this type of solar cell can be easily implemented.

2. Materials and research methodology

Silicon thin films were deposited using a Hybrid Vacuum Deposition System SPT320-PE (Plasmionique, Canada). Kurt Lesker's targets were used for the thin-film deposition. In both cases, the purity of the 2" discs was 99.999%. For a P-type silicon, the dopant was boron, while for N-type silicon (used to produce solar cells), the dopant was phosphorus. Magnetron guns were placed at the top and directed downwards perpendicular to the sample placed on the table. The process was carried out with a gas flow adjusted to 7 sccm and in an argon (Ar) atmosphere at a pressure of $5 \cdot 10^{-3}$ Torr. The magnetron power was set to 75 W. Additionally, the substrate polarization was set to 60 V. The process was carried out in Ar (99.9999% pure) atmosphere. The substrate temperature was controlled using a K-type thermocouple built in Inconel. A proportional-integral-derivative (PID) controller and a thermocouple signal assured feedback control of the sample temperature. Samples were produced with a variable substrate temperature and a variable deposition time. The substrate temperatures selected for deposition were 25 °C, 200 °C, and 400 °C. The maximum deposition time was 120 min. The experiments involved specimens comprising glass plates and silicon wafers coated with a Si thin film through PVD. The dimensions of the substrates were 20 mm × 15 mm. Before deposition, all substrates underwent a thorough cleaning process in an ultrasonic cleaner, consisting of three sequential steps: in water with detergent for 15 min, then in acetone for 10 min, and finally in isopropyl alcohol for 10 min. Finally, it was cleaned using an ion gun. The thicknesses of individual layers in the solar cell were selected based on optical and electrical properties tests. Thin films were deposited on special substrates (S211 Ossila glass) to prepare solar cells. A layer of amorphous silicon with P-type conductivity was deposited on substrates with an ITO layer for 40 min, then a P- and N-type targets were sputtered from two targets at the same time for 40 min to get a P-N junction and, finally, an N-type layer for 40 min. The final stage of the preparation was the deposition of the silver contacts by the MS method, allowing the samples to be tested. A multi-electrode cathode deposition mask was employed to fabricate eight distinct pixels.

The X-ray diffraction research has been conducted on an X'Pert Pro MPD diffractometer by PANalytical, utilizing the strained (Fe filter) radiation of an X-ray tube with a cobalt anode (Co λ = 1.7909 Å), assisted with 40 kV voltage, with a fibre current = 30 mA.

The surface topography of the examined samples was assessed using a Park Systems XE100 atomic force microscope (Park Systems, Suwon, South Korea). The research was implemented in a non-contact mode, on areas of 2 × 2 μm^2. 2D images and their 3D representation were registered. Additionally, fundamental roughness parameters were computed. The vibration frequency of the cantilever was 300 kHz. Registered test results were elaborated in the Park Systems XEI 4.3.1 program.

The thickness of the prepared thin films was determined using a FR-pRo-UV/VIS spectroscopic reflectometer (ThetaMetrisis SA., Peristeri, Greece). The tests were performed in the reflective mode.

The optical properties of the thin films were registered using a UV-VIS 220 Evolution spectrophotometer from Thermo Fisher Scientific Company (Waltham, MA, USA). Research was carried out in the scope of 250–900 nm. Transmittance and absorbance were estimated utilizing an Integrating Sphere Accessory ISA-220.

The electrical properties of ready-made solar cells containing the tested silicon thin films in their structure were examined using an Ossila Solar Cell Testing Kit comprising a source measure unit and an LED-driven solar simulator. The irradiance during measurement was 1000 W/m^2.

3. Results and discussion

Selected results for P-type conductive layers are presented in Figs. 3–9. Figure 3 shows that all X-ray diffraction (XRD) spectra illustrate the broad peak at 2Θ = 19.00–24.00° pointing at amorphous Si (Fig. 3). Its intensity decreases depending on substrate temperature (Ts). This is higher at Ts = 25–200 °C than at Ts = 400 °C. Apart from this broad peak, the XRD spectra of the layers do not exhibit any additional peaks.

The impact of changing the sample temperature on the surface topography of the deposited silicon thin films is shown in Fig. 4. Changing the temperature from room temperature to 200 °C slightly impacted the surface topography [Fig. 4(a) and (b)].

Figure 5 shows the histogram of the unevenness distribution obtained using an Atomic Force Microscopy (AFM). The roughness histogram generated from AFM data represents the distribution of surface height values across the scanned area. A narrow, sharp peak in the histogram suggests a relatively uniform surface with most height values concentrated around a single mean value. A broader or more spread-out histogram indicates a more varied surface with a wide range of height values, reflecting greater roughness. In this case, most of the unevenness of the layer deposited at a temperature of 25 °C does not exceed 2.5 nm, while an increase in temperature to 200 °C

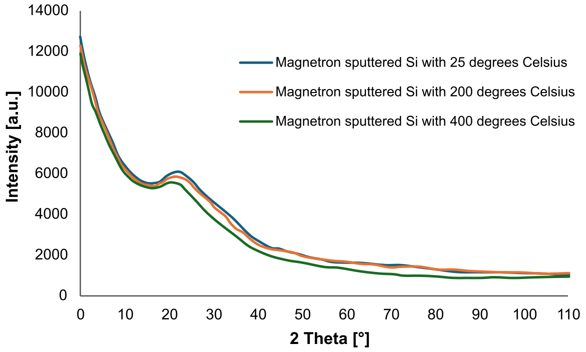

Fig. 3. Diffraction pattern of P-type Si thin films deposited with different substrate temperatures.

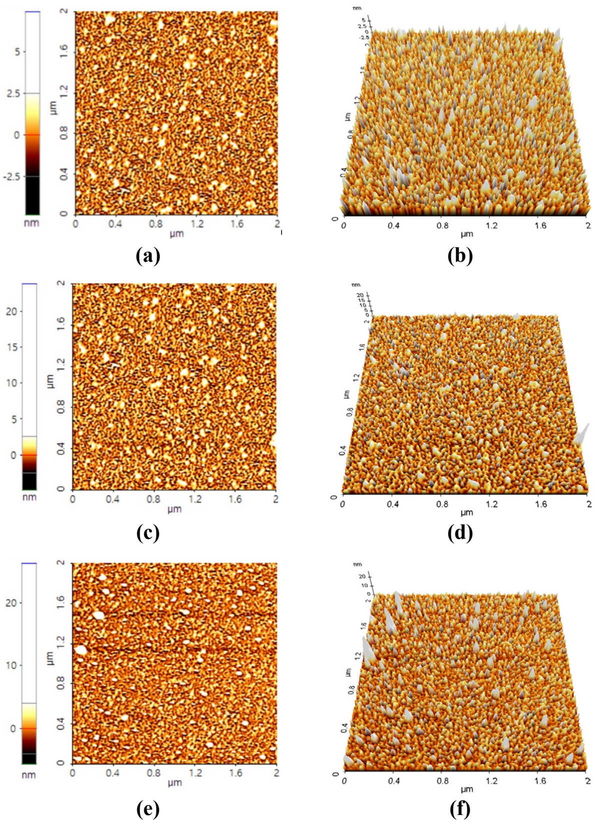

Fig. 4. 2D and 3D AFM images of the surface topography of Si P-type thin film deposited with substrate temperature: 25 °C (a), (b), 200 °C (c), (d), and 400 °C (e), (f).

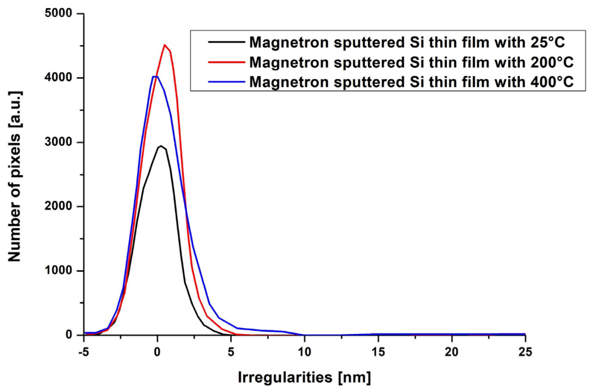

Fig. 5. Histograms of the frequency of surface irregularities of the Si P-type thin films deposited with different sample temperatures.

caused the range to expand to 5.0 nm (Fig. 5). The increase in substrate temperature also resulted in the appearance of sporadic large irregularities reaching up to 23.8 nm (Table 1). Increasing the substrate temperature to 400 °C caused further changes in the surface topography [Fig. 4(e) and (f)] and roughness parameters. The frequency of larger irregularities has increased (Fig. 5), reaching up to 26.2 nm (Table 1).

Table 1
Roughness parameters of the deposited samples.

Substrate temperature [°C]	Ra [nm]	Rq [nm]	Max. irregularity [nm]
25	1.0	1.2	7.4
200	1.1	1.3	23.8
400	1.5	2.1	26.2

Variations in the roughness of deposited layers can occur due to an increased sample temperature which enhances both atomic mobility and diffusion rates. This results in the formation of grains and, subsequently, an increase in grain size, thus changing the roughness. This is associated with the obtained XRD results which showed slight differences in diffraction patterns. Further temperature increases should increase the diffusion rate. Such a situation should cause a reduction in roughness again.

Light reflection within the 250–600 nm range was meticulously measured to determine the thickness of the layers. Figure 6 illustrates an experimental spectrum and its corresponding fitted counterpart, clearly representing the process. All results exhibited a high fit factor, with the R^2 parameter consistently exceeding 0.980 (indicating proximity to the ideal value of 1). The thickness was determined based on these results. The effect of deposition time and substrate temperature on the thickness of the deposited Si thin film is illustrated in Fig. 7. From the obtained data, the growth rate of individual thin films was determined. Consequently, a thin film of Si deposited with a 25 °C substrate temperature is characterised by approximately 1.4 nm/min and decreases with increasing temperature. At a temperature of 400 °C, it is about 1 nm/min. As the sample temperature increases, both the mobility and diffusion rate of the atoms increase which leads to the formation of a densely packed structure affecting the layer thickness. Conversely, according to the theory of a non-spontaneous nucleation, the critical energy for nucleation rises with increasing temperature, thereby decreasing the probability of nucleation. This may lead to a reduced deposition rate of silicon thin films on the substrate.

The transmittance of the glass substrate in the range of 350÷900 nm remains above 90% (Fig. 8). The transmittance of layers thickness ranged up to approximately 90 nm (deposited for up to 90 min) and remained at a high level of over 70%. A further increase in thickness caused a significant decrease in transmittance. For example, a layer with a thickness of approximately 120 nm (deposited in 120 min) has a transmittance in the 30–70% range. A characteristic broadening of the absorbance spectrum was recorded for amorphous silicon thin films compared to pure glass in the range of 350÷400 nm (Fig. 9). The absorbance is stronger as the layer thickness increases.

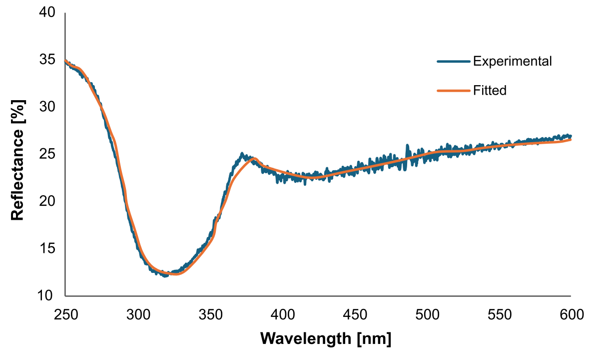

Fig. 6. Example of the experimental and fitted spectrum of a Si P-type thin film deposited with a 400 °C substrate temperature after 60 min.

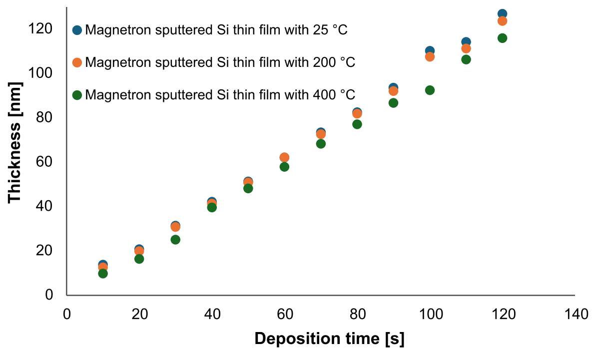

Fig. 7. The influence of the deposition time on the thickness of the prepared Si thin films.

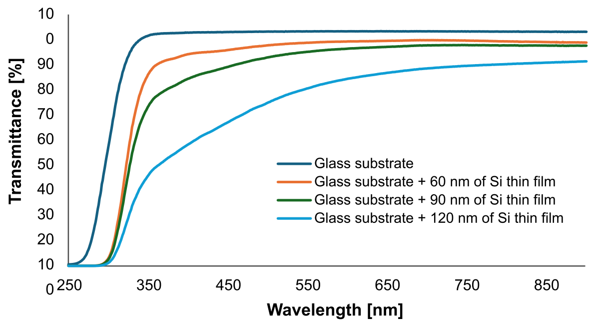

Fig. 8. The influence of a Si P-type thin-film thickness on its transmittance.

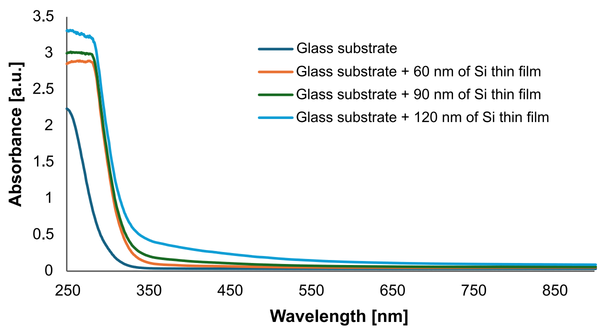

Fig. 9. The influence of a Si P-type thin-film thickness on its absorbance.

Nevertheless, as the light wavelength increases, the absorbance of amorphous silicon decreases, approaching near zero above 650 nm.

Figure 10 shows an example of the current-voltage characteristics of the prepared photovoltaic cells. The irradiance during measurement was 1000 W/m^2. Based on the performed tests, the optimal thickness of the P-type silicon layer in thin-film silicon solar cells was estimated. It depended not only on the obtained test results but also on the remaining layer thicknesses. The best result was presented when the N-type silicon layer was approximately 60 nm. The short circuit current was 12.20 mA cm^{-2}, the open circuit voltage was 0.81 V, and the fill factor was 0.51 (Fig. 10). The average efficiency value was 5.04 %. Further research is planned on optimizing the thickness of silicon layers to increase the short-circuit current and, consequently, improve the final efficiency of the prepared photovoltaic cells.

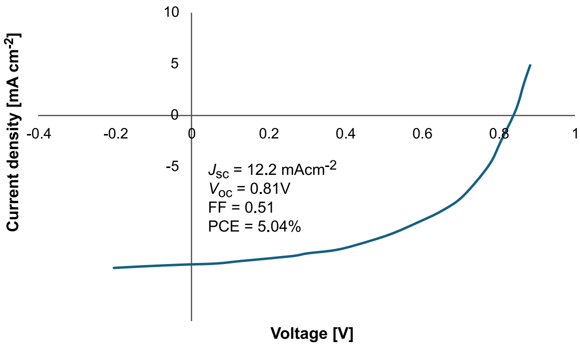

Fig. 10. Light current-voltage characteristics of the produced thin-film silicon solar cell (thickness about: 60 nm P-type, 60 nm P-N junction, 60 nm N-type).

4. Conclusions

The article examined the influence of MS deposition parameters, such as substrate temperature and sputtering time, on silicon layers structure, surface morphologies, and optical properties. Silicon thin films with an amorphous structure were obtained regardless of the parameters used. Small changes in the diffractograms were recorded, requiring further detailed analysis, e.g., TEM. The increase in substrate temperature resulted in an uneven layer growth and the roughness of the prepared layers increased. Changes in the thickness of the deposited layers were recorded depending on the substrate temperature used. This may be related to the mobility and diffusion rate of atoms, which causes changes in the packing density of the layer. It may also be associated with the theory of non-spontaneous nucleation, wherein the critical nucleation energy increases with rising temperature resulting in a decrease in nucleation probability. The high transparency of the thin films was maintained even for a thickness of about 90 nm and was over 80% in the range of 350–900 nm. In order to check the possibility of using deposited layers in photovoltaics, experimental solar cells were manufactured. Based on the obtained I-V characteristics, basic electrical parameters were measured. A photovoltaic cell with a performance of 5.04% was obtained. The obtained results constitute the basis for further work on thin-film silicon photovoltaic cells which are a substitute to bulk silicon photovoltaic cells. Further research is planned to optimize the thickness of individual layers and the impact of additional heat treatment on their structure. The authors applied the MS method to deposit the layers, selecting it instead of the commercially employed CVD method. This replacement could alleviate the necessity for high temperatures and broaden the potential applications of thin-film solar cells.

Authors' statement

Research concept and design, M.S.; collection and/or assembly of data, M.S., M.M.S., K.M., K.L., P.N.; data analysis and interpretation, M.S., M.M.S., P.N.; writing the article, M.S., M.M.S.; critical revision of the article, M.S.,K.M.,K.L.; final approval of article, M.S., K.L.

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