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Electrical and optical properties of silicon-carbon films deposited by electron beam evaporation of silicon carbide in gas environment

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Silicon-carbon films were prepared by electron beam evaporation of silicon carbide in nitrogen, oxygen, helium and propane. Electron beam was produced by electron source with plasma cathode. The resistivity, conductivity activation energy, optical band gap as well as element content of silicon-carbon films obtained at different temperatures were measured. It is shown that a change in the films properties with the deposition temperature correlates with a change in the ratio of elements in the films.

Keywords: plasma-cathode electron source; electron beam evaporation of silicon carbide; silicon-carbon films; resistivity; activation energy; optical band gap.

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Introduction

Silicon-carbon (SiC) films, i.e., films made of silicon carbide containing additional elements such as oxygen, nitrogen, and hydrogen, are used for passivating contacts of silicon solar cells [1, 2] and also as a material for solar cells [3]. This promotes the interest in the electrical properties of such films. Among multiple methods for depositing silicon-carbon films, a specific place is occupied by vacuum methods, and in particular, electron beam evaporation. The advantages of this method compared to traditional plasma-chemical deposition include a relatively high deposition rate (up to several micrometers per minute), ability to deposit at low (down to room) temperatures of a substrate, and absence of toxic organometallic compounds. In papers [4–6], silicon carbide was evaporated using thermionic electron guns, which imposes limitations on both the range of operating pressures and the ability to evaporate material in reactive gases. This paper utilizes an electron source with a plasma cathode [7], which provides

operability in various gases at pressures within the range of units to tens of Pascals. As some papers [8–10] emphasize, the electrical and optical properties of silicon-carbon films are largely determined by deposition conditions. Films deposited by the plasma-chemical deposition method with plasma generated by radio-frequency discharge feature the activation mechanism of electrical conductivity with activation energy $E_a = 0.1\text{--}0.9\text{ eV}$ [8], which increases as the power input into the discharge grows. At the same time, the resistivity also increases within the range of $10^2\text{--}10^9\text{ }\Omega\cdot\text{cm}$. The paper authors report that the increase in power is accompanied by a change in the Si/C atomic concentration ratio from 9 to 2.3, i.e., by elimination of excess silicon. Films deposited by reactive magnetron sputtering of silicon in methane [10] have the resistivity ranging from 1.3×10^3 to $9.4\times 10^9\text{ }\Omega\cdot\text{cm}$ and the activation energy of 0.16 to 1 eV with the methane flow rate increasing. At the same time, the deposition rate decreases from 30 to 12 nm/min, and Si/C atomic concentration ratio also decreases from 2.5 to

0.17. It should also be noted that optical band gap E_g grows from 1.7 to 2.7 eV. Only one paper [4] is devoted to studying the electrical conductivity mechanism of the films deposited by electron beam evaporation. As indicated in paper [4], the film resistivity exhibits non-monotonical dependence on the deposition temperature in the range of 300–575 K, while the activation energy is 0.16 eV.

According to paper [5], the resistivity of thin silicon carbide films generated by electron beam evaporation does not depend on the deposition temperature. Both in [4] and [5], the composition of the gas environment in the vacuum chamber was not controlled. These results call for further studies of the electrical properties of silicon-carbon films vs. the deposition temperature and gas environment composition during electron beam evaporation.

The purpose of this work is to study the electrical and optical properties of silicon-carbon films deposited by the electron beam evaporation of silicon carbide in various gases vs. the deposition temperature.

Experimental technique and methodology

The experiments were conducted using the electron beam evaporation setup described previously in paper [11]. The stainless steel working chamber housed a black-lead crucible with a target sintered from silicon carbide powder, and three substrate holders with individual heaters equipped with thermocouples for measuring the deposition temperature T_{dep} . The substrate holders were shielded by motorized shutters. Optical measurements were performed using SF 2000 spectrophotometer for the films deposited on quartz substrates. Polycore plates sized 15×20 mm and 1-mm thick were used as substrates for electrical measurements. Copper strips 1 mm wide were pre-deposited onto the substrates by vacuum evaporation.

The substrates were placed in the substrate holders, and the working chamber was evacuated using a mechanical pump to a pressure of 1 Pa. After this, the working gas such as nitrogen, oxygen, helium, or propane was injected into the chamber, followed by purging during 10 minutes with simultaneous heating of the substrate holders. At the operating pressure of 4 Pa, the

electron source was activated, and emission current I_e and accelerating voltage U_a were gradually increased until evaporation of the target began. After the operating parameters of $I_e = 90$ mA and $U_a = 11$ kV were reached, the shutters were opened, and films were deposited. The process ended by switching off the electron source. After being cooled, the substrates were removed from the chamber and placed in the thermal evaporation setup, where copper strips were deposited perpendicular to the lower ones. The capacitor structures prepared this way were placed in a temperature-controlled space, and current-voltage characteristics were measured at different temperatures. Current was measured using F 195 instrument (Russia). Film thickness was determined using MII-4 interference microscope (LOMO, Russia). Based on these measurements, the resistivity was calculated. The elemental composition was determined using TM-1000 scanning electron microscope (Hitachi, Japan) equipped with EDS-SWIFT energy-dispersive spectroscopy attachment (Great Britain).

Experiment results and discussion

The current-voltage characteristics of metal–SiC film–metal structures exhibited a linear pattern. The slope of the characteristics changed as the measurement temperature changed. Resistance was determined based on the value of derivative dU/dI at $U = 0$ [12]. The curves of resistivity ρ of the films deposited in different gases (Fig. 1) show a decrease of ρ as the deposition temperature increased. Moreover, the highest resistivity was demonstrated by the films deposited in oxygen and propane.

The logarithm of film resistance features the linear dependence on the reciprocal temperature, thus indicating the conductivity activation mechanism. Conductivity activation energies E_a , determined from the slope of the straight lines are shown in Fig. 2a. As is seen, the values of E_a also decrease as the deposition temperature increases. The nature of the derived curves correlates with the behavior of optical band gap E_g , derived from processing transmittance spectra in the visible region using Tauc coordinates [13] (Fig. 2b). It is worth noting that the values of E_a and E_g differ significantly.

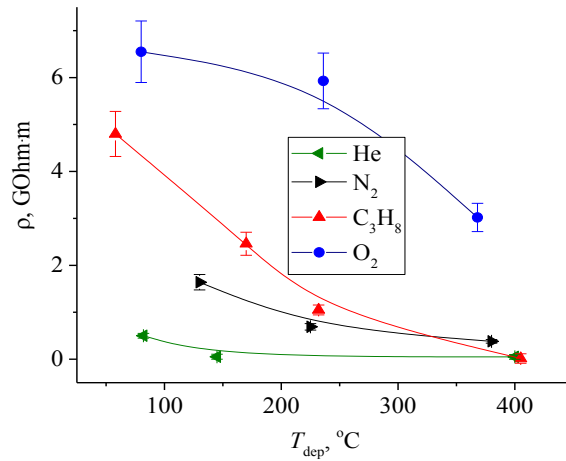


Fig. 1. Resistivity of the films deposited in different gases vs. deposition temperature T_{de}

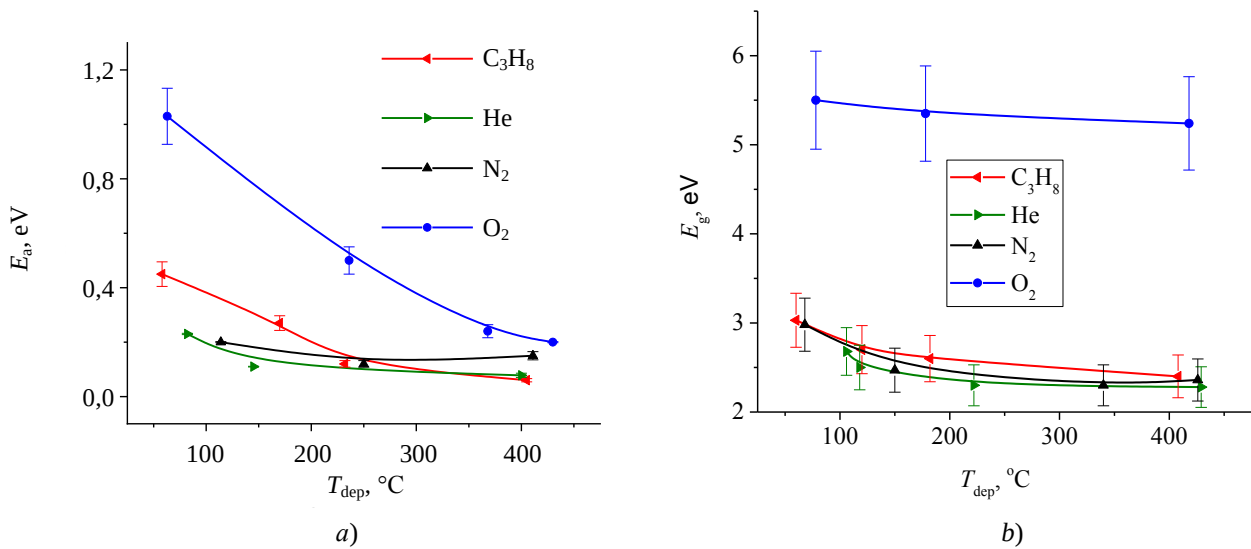


Fig. 2. Values of activation energy E_a (a) and optical band gap E_g vs. deposition temperature T_{dep} in different gases

Parallel measurements of film composition allow for some assumptions regarding the reasons for the change in resistivity depending on the deposition temperature. Temperature increase causes a rise in the relative silicon content (Fig. 3). This result agrees with the data presented in papers [4, 6] where decreased atomic concentration ratio Si/C is recognized as the primary cause for the increase in E_g and E_a values. It should be noted, however, that in our case, oxygen plays a certain role. Its content changes only slightly as the deposition temperature increases; consequently, the Si/O ratio increases as well with the rising deposition temperature. The reason for the increased silicon content with temperature is likely due to a faster decrease in the adsorption of gas molecules and carbon atoms compared to silicon atoms. An argument in favor of this assumption is the higher adsorption energy of carbon compared to

silicon, and therefore a stronger dependence of its adsorption on temperature [14]. Furthermore, since carbon monoxide is a gas, the interaction of carbon with oxygen as the temperature grows also contributes to decreased carbon content in the film, as the gas is carried away into the pumping system. Resistivity values of the deposited films allow them to be classified as semiconductor materials. The relatively low activation energy indicates that the film material has properties differing significantly from those of crystalline silicon carbide. This is also evident during the analysis of optical transmittance spectra. The E_g value determined from these spectra and lying within 2–3 eV (Fig. 2b) is in reasonable agreement with the values of this parameter for crystalline SiC. The difference observed between the values of E_g and E_a can be explained by the fact that donor-type impurity levels [15] are assumed to be present within the band gap of the

films, with a depth corresponding to the measured E_a values. These levels can be caused both by impurities, namely hydrogen and oxygen, and by the amorphous structure of the film. The latter

assumption is quite probable because the known papers [8–10] devoted to the electrical properties of silicon-carbon films report E_a values equaling to the deciles of electronvolt.

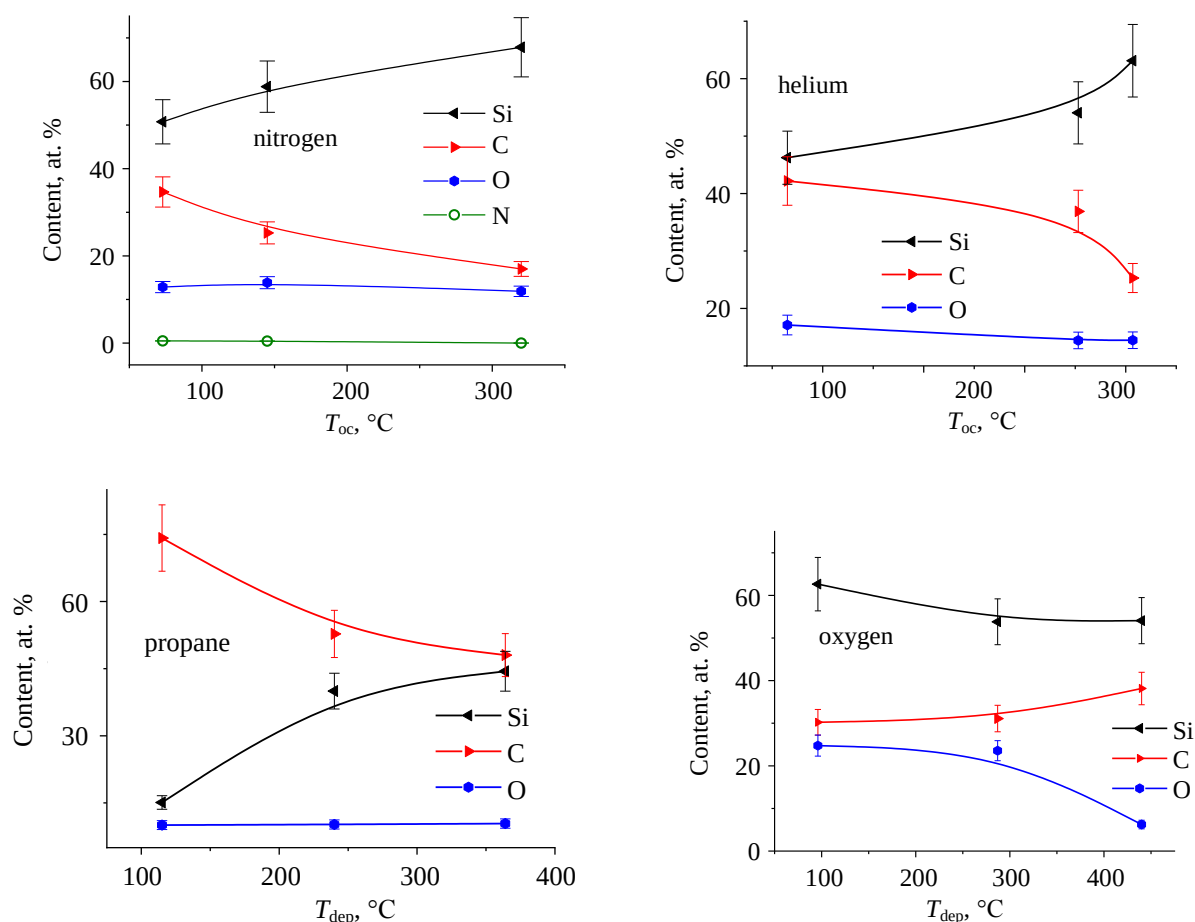


Fig. 3. Composition of the films deposited in different gases vs. deposition temperature T_{dep}

Conclusion

The measurements performed are indicative of the conductivity activation mechanism for silicon-carbon films produced by electron beam evaporation of silicon carbide in different gases. The activation energy value ranges from 1.0–0.1 eV depending on the gas type and decreases as the deposition temperature increases. The resistivity of the films is 10^8 – 10^9 $\Omega\cdot\text{m}$ and tends to decrease with the increasing deposition

temperature. The observed regularities are explained within the framework of concepts about the change in film composition towards silicon enrichment with the increasing deposition temperature.

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