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Comparative characteristics of $TiO_2(Er_2O_3, Dy_2O_3)$/por-SiC/SiC heterostructures (Review)

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Abstract. In this work, comparative characteristics of thin oxide films (OF) of titanium, erbium, and dysprosium formed on silicon carbide substrates in the presence and absence of a porous silicon carbide (por-SiC) layer have been considered. It has been shown that regardless of the presence of a porous buffer layer in the $TiO_2(Er_2O_3, Dy_2O_3)$/por-SiC/SiC and $TiO_2(Er_2O_3, Dy_2O_3)$/SiC structures, oxide layers of the approximately equal thickness are formed, and quality of the interface in OF/SiC structures is higher than that in the OF/por-SiC/SiC structures. An increase in the time and temperature of rapid thermal annealing makes it possible to improve the quality of the oxide film/substrate interface regardless of the presence of a porous buffer layer in the structure. In this case, the narrowest interface "oxide film/buffer porous layer/substrate" is observed for the TiO_2 /por-SiC/SiC structures. The TiO_2 /por-SiC/SiC structures are most sensitive to changes in the parameters of rapid thermal annealing, and the Er_2O_3/por-SiC/SiC structures are the most stable.

Keywords: interface, thin oxide films, porous silicon carbide, Auger spectrometry, optical absorption, photoluminescence.

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1. Introduction

Nowadays, in the literature, oxides of rare earth elements (REE) are often considered as alternative oxides used in oxide/semiconductor structures [1-5]. These oxides possess high transparency in the visible spectral range, chemical and thermal stability, and have an optimal refractive index for these purposes [1, 2].

This interest in the search for alternative oxides is caused by the fact that a decrease in the scale of MIS devices is accompanied by a decrease in the channel length and thickness of the gate dielectrics [5]. Decreasing in the leakage current through the gate dielectric is achieved due to the use of the so-called alternative dielectrics with high dielectric permittivity – high-*k* dielectrics [5].

Use of high-*k* dielectrics allows increasing the physical thickness of the dielectrics and thus suppressing the tunneling current [5, 6]. Another application of high-*k* dielectrics is associated with their use in the storage capacitor of the random access memory cells [6]. Recently, it was proposed to use the high-*k* dielectrics to increase the performance of FLASH memory [6].

Direct formation of REE oxides on the surface of a semiconductor substrate is difficult because of the high coefficient of interdiffusion [3]. Moreover, the structural, optical and electrical characteristics of oxide films often critically depend on the methods and conditions of preparation, on subsequent processing, and also on the type of substrates used [2, 4, 7-9]. One way to reduce this effect is to create a porous layer between the substrate and the epitaxial layer [10, 11].

In this work, comparative studies of titanium, erbium, and dysprosium oxide films (OF) formed on silicon carbide substrates in the presence and absence of a porous silicon carbide (por-SiC) buffer layer have been performed.

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2. Discussion of the experimental data

2.1. Features of formation of the interface in the $TiO_2(Er_2O_3, Dy_2O_3)$/por-SiC/SiC heterostructures

As it was shown in [12, 13], the use of rapid thermal annealing (RTA) makes it possible to reduce the thickness of the oxide film/substrate interface in the $TiO_2(Er_2O_3, Gd_2O_3)$/SiC structures. RTA has a similar effect on the thickness of the interface in the $TiO_2(Er_2O_3, Dy_2O_3)$/por-SiC/SiC structures [14-16].

Fig. 1 shows the atomic profiles of the TiO_2/por-SiC/SiC, Er_2O_3/por-SiC/SiC and Dy_2O_3/por-SiC/SiC structures near the interface during various heat processing [14-16].

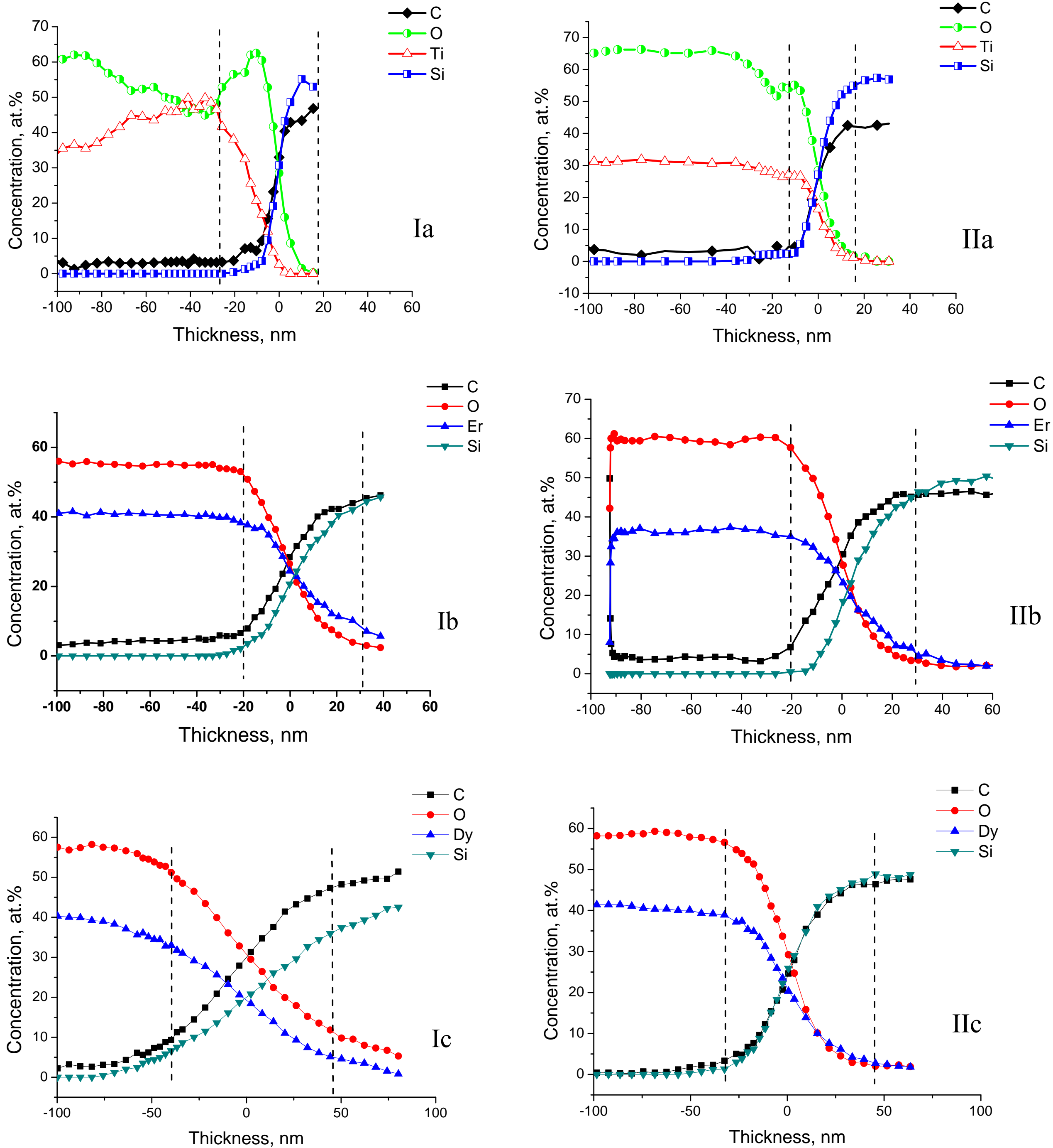

Fig. 1. Atomic profiles of distribution of the components in the TiO_2/por-SiC/SiC [15] (*a*), Er_2O_3/por-SiC/SiC [14] (*b*) and Dy_2O_3/por-SiC/SiC [16] (*c*) structures at the RTA temperature 900 °C (I) and 1000 °C (II), respectively. The RTA time is 1 s (*1*) and 5 s (*2*). The vertical lines indicate the area of the interface.

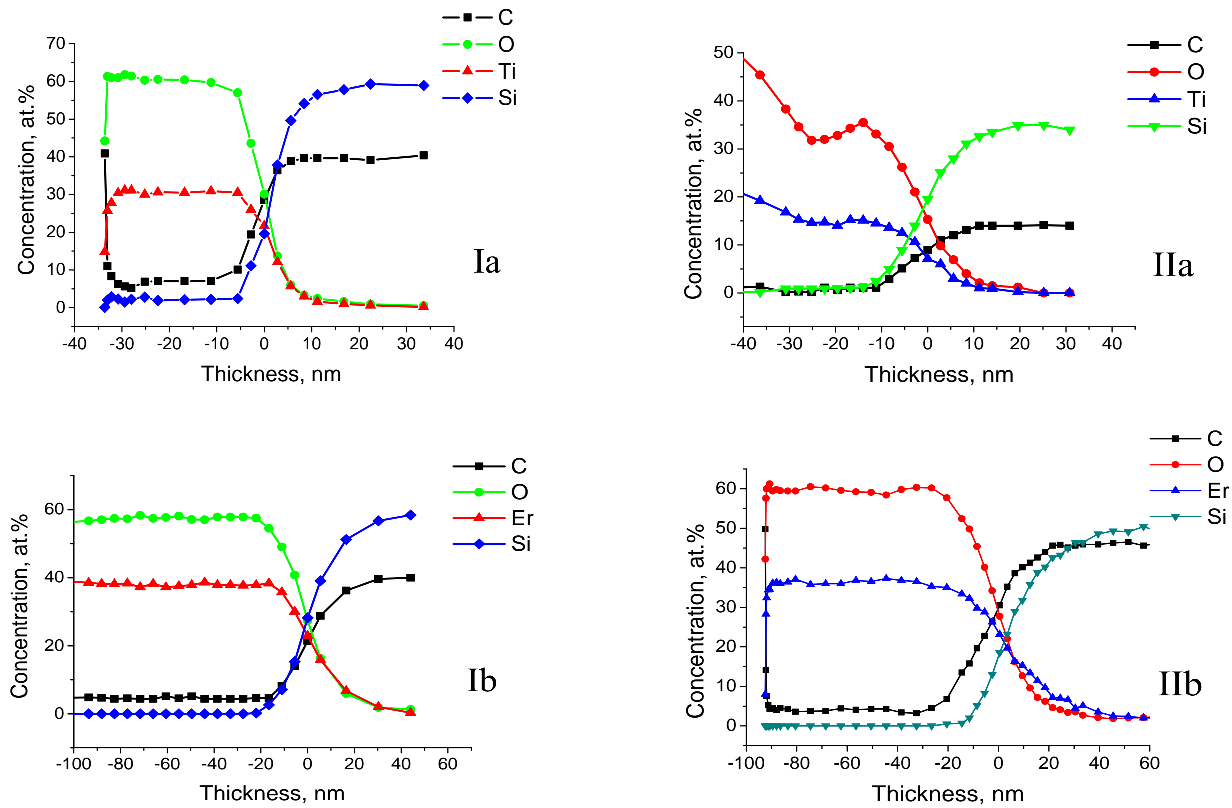

Fig. 2. The contents of elements in atomic percents in the TiO_2/SiC (Ia) and TiO_2/por-SiC/SiC (Ib) structures as well as in the Er_2O_3/SiC (IIa) and Er_2O_3/por-SiC/SiC (IIb) structures [12, 14, 15, 18].

For the TiO_2/por-SiC/SiC structures, qualitative estimations based on experimental data on Auger profiling [15] have shown that vacuum heat processing of thin metal titanium films deposited on a por-SiC/SiC substrate initiated the appearance of at least a two-layer system: the upper layer was formed by the TiO phase, under which there was a more extended area with a phase composition close to TiO_2.

A distinctive feature of high-temperature annealing in vacuum is formation of a layer heterogeneous in thickness at the interface because of creating TiC [17, 18]. Rapid thermal annealing at the temperature 900 °C leads to formation of an oxide titanium film of variable composition (Fig. 1a). An increase in the temperature of RTA stabilizes the phase composition of the formed titanium oxide over the entire thickness (Fig. 1a). The phase composition of the obtained oxidized layers corresponds to the structure of rutile, the ratio of the components of TiO_2 oxide is $N_O/N_{Ti} \approx 2.2$.

As it follows from the Auger spectrometry data, during the growth of erbium and dysprosium oxides, heat processing leads to formation of oxide layers of Er_2O_3 and Dy_2O_3 uniform in thickness, their composition is close to the stoichiometric one (Figs 1b, 1c). The ratio of the components of Er and Dy oxides formed on the SiC substrate in the presence of a buffer por-SiC layer practically corresponds to the stoichiometric composition of one and a half oxide: $N_O/N_{Er} \approx 1.3\ldots1.6$ for Er_2O_3 and $N_O/N_{Dy} \approx 1.4$ for Dy_2O_3, regardless of the oxidation time.

The observed changes in the composition of oxide phases in the near-boundary layers and their extent are related with the conditions for formation of oxide films. As can be seen from Fig. 1, the chemical composition of the transition areas "oxide film/substrate" is different from that of the bulk of oxides. An increase in the time of RTA leads to formation of a sharper interface "oxide film/substrate" [13, 14, 16, 19, 20].

It is caused by the fact that an increase in the RTA time leads to an increase in the fraction of the oxide film with a stoichiometric composition with respect to the underoxidized metal layer, as well as to destruction of metal silicates [21] and metal carbides [17, 18] formed in the intermediate layer "oxide film/porous layer". The thickness of the interface of TiO_2(Er_2O_3, Dy_2O_3)/por-SiC/SiC structure depends on the chemical composition of oxide. So, the narrowest interface "oxide film/ substrate" is observed for TiO_2/por-SiC/SiC structures; the interface thickness is ~ 30 nm (Fig. 1 (IIa)).

For the Er_2O_3/por-SiC/SiC and Dy_2O_3/por-SiC/SiC structures, this value is ~ 50 nm (Fig. 1 (IIb)) and ~ 76 nm (Fig. 1 (IIc)), respectively. In this case, the TiO_2/por-SiC/SiC structures are most sensitive to additional heat processing. An increase in the RTA temperature in this case leads to a decrease in the interface thickness by ~ 25 nm (Fig. 1a). The Er_2O_3/por-SiC/SiC structures turned out to be the least sensitive to changes in the RTA parameters. In this case, the interface thickness was found to be practically unchanged (Fig. 1b).

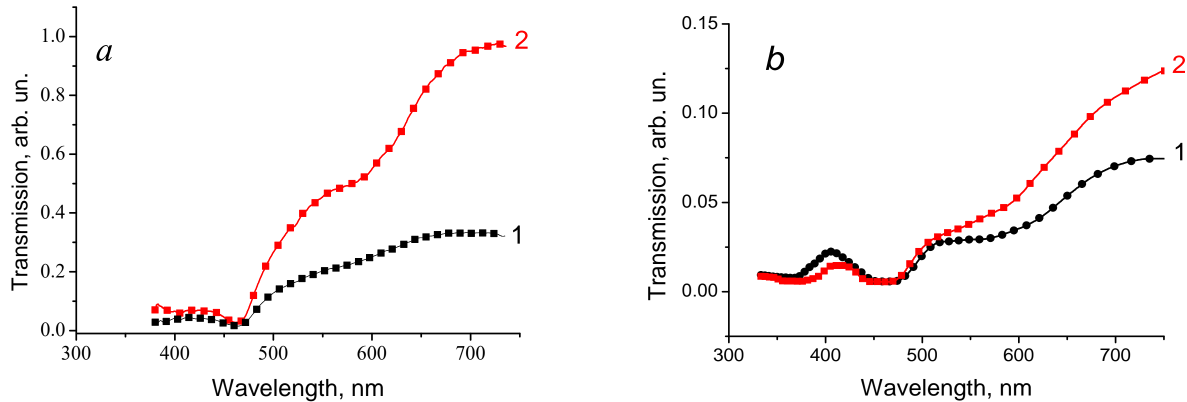

Fig. 3. Transmission spectra of the Er_2O_3/por-SiC/SiC (*a*) and Dy_2O_3/por-SiC/SiC (*b*) structures. The RTA time is 1 s (*1*) and 5 s (*2*) [14, 16].

Since porous silicon carbide layers are often used in complex structures to reduce the concentration of defects at the interface between a porous layer and an epitaxial film [10, 11, 22], it is of interest to compare the characteristics of oxide film/SiC structures in the presence and absence of a buffer layer of porous silicon carbide between the oxide film and the silicon carbide substrate.

A comparison of the thickness of oxide layers determined using the Auger spectrometry method showed that both in the presence of a buffer layer and in the absence of it, layers of approximately the same thickness close to 60 nm in the case of titanium oxide [15] and 80…100 nm in the case of erbium oxide [12] were formed.

Fig. 2 shows the atomic profiles of the heterostructures formed by titanium and erbium oxides on the SiC substrate in the absence and presence of a buffer por-SiC layer at the oxide-semiconductor interface [12, 14, 15, 18].

As can be seen from Fig. 2, the ratio of the components for titanium and erbium oxides formed both directly on the SiC crystalline substrate and in the presence of a buffer por-SiC layer is close to the stoichiometric composition typical for these oxides. For titanium oxide, this ratio is $N_O/N_{Ti} \approx 1.98$ for oxides formed directly on the SiC crystalline substrate and $N_O/N_{Ti} \approx 2.2$ for oxides formed on SiC substrate in the presence of a buffer por-SiC layer, which corresponds to the modification of titanium dioxide with the rutile structure. In the case of erbium oxide: $N_O/N_{Er} \approx 1.6…1.7$ and $N_O/N_{Er} \approx$ 1.3…1.6 for oxides formed on a SiC substrate in the absence and presence of a buffer por-SiC layer, respectively.

At the same time, the chemical composition of the interface “oxide film/substrate” differs from the chemical compositions of the oxide and substrate (Fig. 2). In this case, a sharper interface “oxide film/substrate” is observed, when TiO_2 and Er_2O_3 oxides are formed directly on the crystalline substrate.

2.2. Transmission and photoluminescence spectra of the $TiO_2(Er_2O_3, Dy_2O_3)$/por-SiC/SiC heterostructures

Figs 3a, 3b shows the transmission spectra of the Er_2O_3/por-SiC/SiC and Dy_2O_3/por-SiC/SiC structures. As can be seen from Fig. 3, an increase in the RTA time, like to the TiO_2/por-SiC/SiC structures [13, 14], leads to an increase in transmission in the Er_2O_3/por-SiC/SiC structures [12, 19, 20] and Dy_2O_3/por-SiC/SiC [16] within the spectral range 400…800 nm.

The minimum in the transmission spectrum of the structures is caused by the presence of a nitrogen impurity in the SiC substrate. The increase in the transmission of the OF/por-SiC/SiC structures as well as the decrease in the thickness of the transition layer at the oxide film/substrate interface (Fig. 1), observed with an increase in the RTA time of these structures, are most likely caused by the same reasons. These reasons are as follows: an increase in the fraction of an oxide film with a stoichiometric composition with respect to the underoxidized metal layer [14], as well as destruction of metal silicates [21] and metal carbides [17, 18] at the oxide film/substrate interface.

An increase in the RTA time of OF/por-SiC/SiC structures leads to an increase in the optical transmission of the structure regardless of the chemical composition of the oxide film. However, with the same tendency toward increased transmission, the degree of transparency of Er_2O_3/por-SiC/SiC structures in the range 600…700 nm is almost 7 times higher than that for Dy_2O_3/por-SiC/SiC structures (see Fig. 3). This effect can be explained by the antireflection properties of erbium oxide. So, it was shown in [1, 2] that using the antireflection film of the erbium oxide makes it possible to increase the integrated sensitivity of a silicon photoelectric converter by 30–32% in the wavelength range of radiation from 400 up to 1100 nm.

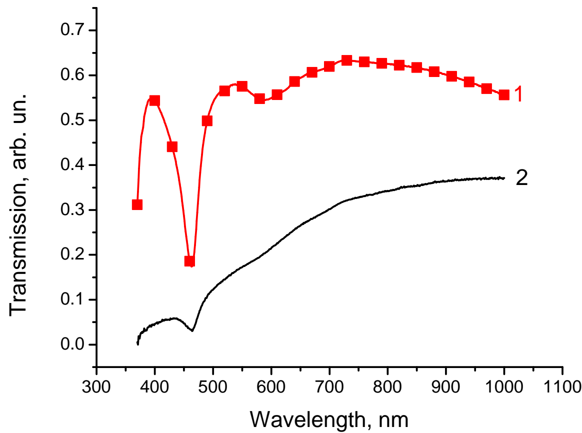

Fig. 4. Characteristic transmission spectra for the structures in which an oxide film is formed directly on a silicon carbide substrate (*1*) and in the presence of a porous buffer layer (*2*).

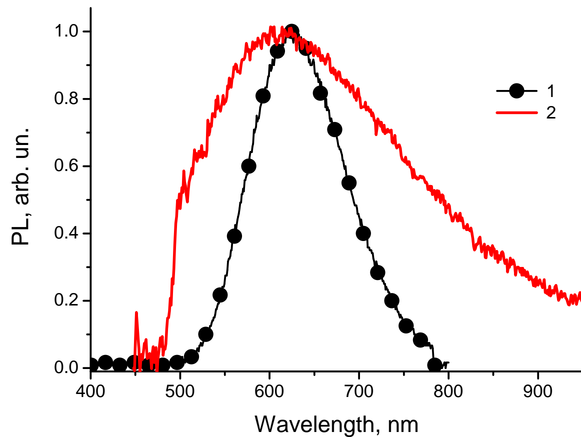

Fig. 5. Characteristic PL spectra for the OF/SiC (*1*) and OF/por-SiC/SiC (*2*) structures normalized to the band maximum [14].

Fig. 4 shows the characteristic transmission spectra for the structures in which an oxide film is formed directly on a silicon carbide substrate and in the presence of a porous buffer layer.

As can be seen from Fig. 4, the OF/por-SiC/SiC structures are characterized by a sharp decrease in optical transmittance as compared with the oxide film/SiC structures, which is caused by the presence of a porous layer and occurs due to an increase in the scattering of porous layer [14, 16, 19].

Fig. 5 shows the characteristic PL spectra of the OF/SiC and OF/por-SiC/SiC structures normalized to the band maximum [14]. It should be noted that the photoluminescence (PL) spectra typical for the structures formed on the same substrates do not depend on the RTA time and are almost identical, however, they change very strongly with a change in the substrate structure.

As can be seen from Fig. 5, in the PL spectra of both OF/SiC and OF/por-SiC/SiC structures, a broad band with a maximum located near 630 nm caused by luminescence of silicon carbide is observed. In the literature [23, 24], this PL band is related to radiative transitions at the centers “impurity (nitrogen) – defect” or “impurity – vacancy” (usually a carbon vacancy is considered). In [25], this band is related with recombination processes caused by the presence of defects near the surface of a silicon carbide crystal. Apparently, just the presence of defects at the OF/por-SiC interface is responsible for a substantial broadening of the PL band in the OF/por-SiC/SiC structures.

In addition, as already noted in [19], the appearance of a broad PL band of por-SiC is related with impurity states that are formed on the por-SiC surface during formation of pores and due to the products of chemical reactions in the course of SiC etching [26-28]. According to [26-28], one of the reasons for PL appearance in por-SiC is surface states caused by the presence of impurities, surface defects, complex compounds (such as oxides and siloxenes) or saturation of Si–H or C–H bonds. When analyzing the PL spectra of multilayer structures, it should be taken into account that the integrated PL spectrum of this structure will be caused by the contribution of both the PL spectrum of the oxide film and PL spectrum of the substrate.

In this case, the characteristics of the substrate PL spectrum will also depend on the transmission spectrum of the oxide film in the range of exciting radiation and in the range of luminescence of the substrate. As it was noted in [12, 14, 16, 18], an increase in the degree of transparency typical for oxide films with increasing the RTA time does not lead to a change in the spectral characteristics of PL structures. Thus, the analysis of the PL spectra of the OF/SiC and OF/por-Si/SiC structures showed that the spectral composition of the PL band of these structures is mainly caused by contribution of the substrate [12, 14, 29].

The broadening of PL band inherent to the structure containing the por-SiC layer as compared with the PL band of the OF/SiC structure is caused by both radiative recombination of impurity states directly in the buffer por-SiC layer [26–28] and the presence of defects at the OF/por-SiC interface [14]. The PL data correlate well with the Auger spectroscopy ones attesting to the blurring of the oxide film/substrate interface in the presence of a porous buffer layer (Fig. 2).

3. Conclusions

Thus, regardless of the presence of a porous buffer layer, the TiO_2, Er_2O_3 and Dy_2O_3 oxide layers of approximately the same thickness are formed in the OF/por-SiC/SiC and OF/SiC structures. However, the presence of the phases of metal carbides or metal silicides formed at the OF/por-SiC interface in the OF/por-SiC/SiC structures leads to

blurring and increasing the thickness of the oxide film/substrate interface. In this case, an increase in the time or temperature of RTA allows creating oxide layers of TiO_2, Er_2O_3 and Dy_2O_3 uniform in thickness, composition that is close to stoichiometric ones.

The observed changes in the composition of oxide phases in the near-boundary layers and their extent are related with different modes of RTA during formation of oxides and their chemical composition. So, the narrowest oxide film/porous layer/substrate interface is observed for TiO_2/por-SiC/SiC structures, the interface thickness is ~ 30 nm. For the Er_2O_3/por-SiC/SiC and Dy_2O_3/por-SiC/SiC structures, this value is ~ 50 nm and ~ 76 nm, respectively. In this case, the TiO_2/por-SiC/SiC structures are the most sensitive to changes in the RTA conditions, and the Er_2O_3/por-SiC/SiC structures are the least sensitive.

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Порівняльні характеристики гетероструктур $TiO_2(Er_2O_3, Dy_2O_3)$/por-SiC/SiC (Огляд)

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Анотація. У даній роботі розглянуто порівняльні характеристики тонких оксидних плівок (OF) титану, ербію та диспрозію, сформованих на підкладках карбіду кремнію при наявності та відсутності пористого шару por-SiC. Показано, що незалежно від наявності буферного пористого шару в структурах $TiO_2(Er_2O_3, Dy_2O_3)$/por-SiC/SiC та $TiO_2(Er_2O_3, Dy_2O_3)$/SiC формуються оксидні шари приблизно однакової товщини, а якість межі поділу в структурах OF/SiC вища, ніж у структурах OF/por-SiC/SiC. Збільшення часу і температури швидкого термічного відпалу дозволяє поліпшити якість межі поділу оксидна плівка/підкладка незалежно від наявності в структурі пористого буферного шару. При цьому найбільш вузька межа поділу оксидна плівка/буферний пористий шар/підкладка спостерігається у структурах TiO_2/por-SiC/SiC. Найбільш чутливими до зміни параметрів швидкого термічного відпалу є структури TiO_2/por-SiC/SiC, а найбільш стійкими – структури Er_2O_3/por-SiC/SiC.

Ключові слова: межа поділу, тонкі оксидні плівки, карбід кремнію, пористий карбід кремнію, Оже-спектрометрія, оптичне поглинання, фотолюмінесценція.