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Influence of Nickel Impurity on the Operating Parameters of a Silicon Solar Cell

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Abstract—The research results present the influence of nickel impurities introduced by diffusion into monocrystalline silicon on the characteristics of solar cells (SCs). It is established that doping with nickel atoms makes it possible to increase the lifetime of the MCCs in the material by up to a factor of two and the efficiency of SCs by 20–25%. It is shown that the distribution of nickel clusters in the volume of the material is almost uniform, and their size does not exceed 0.5 μm. The concentration of clusters in the volume is ~10^{11}–10^{13} cm^{-3}; and in the near-surface layer, ~10^{13}–10^{15} cm^{-3}. The physical mechanisms of the influence of the bulk and near-surface clusters of nickel atoms on the efficiency of silicon SCs are revealed. It is experimentally established that the decisive role in increasing their efficiency is played by the processes of gettering of recombination-active technological impurities by nickel clusters, occurring in the nickel-enriched front surface region of the SCs.

Keywords: silicon solar cell, diffusion, nickel clusters, recombination centers, gettering

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1. INTRODUCTION

In modern industrial production, there is a tendency to increase the share of solar cells (SCs) based on solar silicon, which is due to its relative cheapness [1]. However, solar silicon has a shorter lifetime of minority charge carriers (MCCs) due to the presence in it of a fairly high content of technological impurities (Fe, Cu, Au, Cr, etc.), which makes it difficult to obtain efficient SCs based on it [2].

To increase the efficiency of SCs based on solar silicon, it is necessary, first of all, to increase the lifetime of photogenerated charge carriers (τ) [3], as well as reduce optical and electrical energy losses [4, 5]. The τ of MCCs in SCs can be increased by gettering uncontrolled impurity atoms. The introduction of clusters of nickel atoms in the material is a promising method of doing this [6–9]. It is well known [10, 11] that the distribution of the concentration of nickel atoms after diffusion has significant heterogeneity—a high near-surface concentration—which quickly decreases by depth, and its relatively constant value in the volume.

The works [12, 13] show that diffusion doping with nickel atoms from the front side of an SC with a deep p–n transition leads to an increase in its efficiency. However, at present, the physical mechanisms of the influence of nickel atom clusters, including those located in the near-surface region, on the operational parameters of silicon SCs are unclear. Studying the influence of clusters of impurity nickel atoms on the parameters of silicon SC is of great practical interest due to the technological features of nickel alloying [14–17]. The aim of this study is to establish the influence of nickel atoms located both in the bulk and in the near-surface enriched layer on the efficiency of silicon SCs.

2. EXPERIMENTAL METHODS

For the research, we used polished silicon wafers of monocrystalline silicon grown using the Czochralski method of the KDB-0.5 brand, which is 380 microns thick. The resistivity was 0.5 Ohm cm, the oxygen content was (N_O) ~ 7×10^{17} cm^{-3}, dislocation density was not more than 10^2 cm^{-2}, and MCC lifetime $\tau > 6$ μs. A layer of pure nickel 1 micron thick was deposited in a vacuum. Nickel diffusion was carried out in the temperature range $T_{diff} = 750$–1250°C for $t = 30$ min in air. After diffusion, to activate the gettering process of uncontrolled recombination-active impurities, additional thermal annealing was carried out in air in the temperature range $T_{anneal} = 600$–1100°C for $t = 30$ min [18–20]. After each technological operation, the surface was cleaned and the wafers were chemically

treated (10% HCl, 10% HF) to remove the residual nickel and silicon oxide from the surface.

The distribution of nickel atoms in the surface layer of silicon was measured with a CAMECA IMS-6f Magnetic Sector SIMS mass spectrometer (SIMS). The elemental composition of nickel clusters both on the surface and on the sample's chip was studied using a TESCAN MIRA3 scanning electron microscope in the mode of X-ray local probe microanalysis. The cleavage of the samples was scanned with a step of 0.5 μm, starting from the front side (nickel-doped side).

To study the influence of nickel atoms on the parameters of SCs ($J_{\text{short circuit}}$ is the short circuit current density, U_{xx} is the open circuit voltage, ξ is the filling factor of the volt-ampere characteristic (VAC), $P_{\max}$ is the maximum output power) and lifetime of the MCCs, SCs were manufactured in which the p–n-transition was created by the diffusion of phosphorus into KDB 0.5 silicon wafers of the p-type at $T_{\text{diff}} = 1000$°C for $t =$ 0.5 hours. The depth of the p–n-transition was 0.5 μm. Then the plates were cut into individual samples measuring 1 × 1 cm^2. For the research, three SC groups were formed: Group I, the control group; Group II, a thin layer of pure nickel 1 μm thick, which was deposited in a vacuum on the back side of the plate after phosphorus diffusion; and Group III, a thin layer of pure nickel 1 μm thick, which was deposited in a vacuum on the front side. The diffusion of nickel was carried out in the temperature range $T_{\text{diff}} = 750$–1250°C for t ranging from 3 min to 30 min. After the diffusion of phosphorus and nickel, the back layer of the plate, enriched with nickel and phosphorus, was removed by grinding to a depth of ~10 μm. Additional gettering thermal annealing was carried out at $T_{\text{anneal}} = 750$–800°C. The VACs and lifetime of the nonconductive charge in the SC base were measured after the formation of the nickel contact. There was no antireflective coating on the surface of the elements. The lifetime of the MCCs in the resulting structures was measured by the method described in [21].

For identifying the relative contributions of the surface and bulk nickel atoms, the parameters of the SC were additionally studied by samples in which nickel diffusion was carried out at $T_{\text{diff}} = 850$°C for $t = 30$ min before the formation of the p–n-transition (in this case, the thickness of the near-surface enriched layer is estimated at 2–2.5 μm [11, 13]). After the diffusion of nickel from the front surface of the samples, the Ni-enriched surface layer up to 5 μm thick was removed by polishing. Then the p–n transition was formed according to the technology described above and additional thermal annealing was carried out at $T_{\text{anneal}} = 750$–800°C for $t = 30$ min (sample group IV).

3. EXPERIMENTAL RESULTS

Studies of the nickel distribution after diffusion have shown that the near-surface concentration of nickel can reach $n_S \sim 4 \times 10^{21}$ cm^{-3} and the thickness of the enriched *Ni* layer is $d \sim 3.25$ μm (Fig. 1). An enriched layer with a concentration of the order of magnitude $n_S \sim 10^{20}$ cm^{-3} also appears on the backside; however, its thickness is small ($d \sim 0.5$ μm). In the volume of the samples, nickel is distributed almost uniformly (with the concentration depending on the diffusion mode $n_{Ni} \approx 10^{16}$–2×10^{18} cm^{-3}). In this case, the front surface of the samples, according to the local X-ray probe microanalysis, contains 96.58 at. % silicon, 1.44 at. % nickel, and 1.98 at. % oxygen (Fig. 2). This allows us to state that there are no continuous films of nickel silicides on the surface. It has also been experimentally established that the Ni-enriched layer is preserved during subsequent heat treatments with temperatures below 900°C, and the maximum concentration of nickel atoms in the near-surface regions is practically independent of the diffusion time and weakly depends on the nickel diffusion temperature.

Studies using secondary ion mass spectrometry have shown that diffusion-introduced nickel is distributed in silicon in the form of clusters (Fig. 3). In this case, the average surface concentration of Ni atoms is ~6×10^{14} at./cm^2. According to the electron and IR microscopy data (Fig. 4), it has been established that the surface density of nickel clusters is ~5×10^6–10^7 cm^{-2} on the front surface of silicon and ~$(4$–$5) \times 10^6$ cm^{-2} in the volume (measured on the sample chip). The distribution of clusters in the volume is almost uniform and their size does not exceed 0.5 μm (approximately, 20–200 nm), this is consistent with the results in [22–25]. The concentrations of clusters in the sample volume calculated based on the obtained data are $n_c \approx 10^{11}$–10^{13} cm^{-3}; and in the near-surface region, $n_c \approx 10^{13}$–10^{15} cm^{-3}.

To determine the thermal stability of nickel clusters, additional thermal annealing was performed. It was established that when $T_{\text{anneal}} = 650$–800°C, there is an increase in the size of nickel clusters, and at $T_{\text{anneal}} >$ 900°C, the clusters disappear; i.e., the clusters are decaying. Measuring the composition of the clusters before heat treatments showed that the clusters on the silicon surface consist mainly of silicon atoms (84.93%) and nickel (13.38%); and they also contain uncontrolled impurities of Cu, Fe, and Cr atoms (Fig. 5). After additional heat treatment ($T_{\text{anneal}} = 800$°C, $t = 30$ min), the nickel concentration in the cluster increases to ~40–60%, oxygen up to ~30–35% and rapidly diffusing impurities (RDIs) up to ~30–50% relative to their concentrations before thermal annealing. The results obtained give grounds to state the effective gettering of technological impurities by nickel clusters, which act as recombination centers in silicon. The consequence of this should be a significant increase in the lifetime of the MCCs, primarily due to the formation of high concentrations of nickel clusters in the near-surface layers.

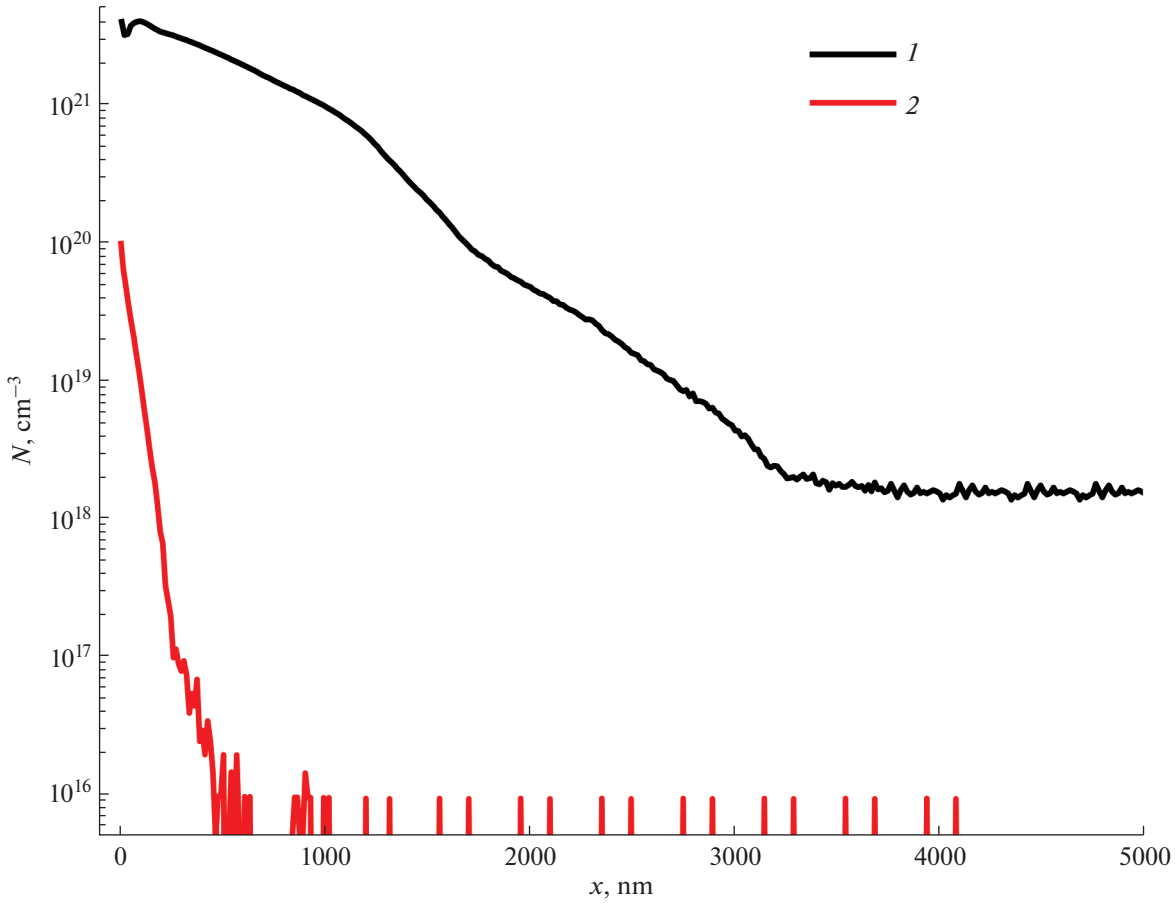


Fig. 1. Distribution of nickel atoms in the front (*1*) and back (*2*) layers of silicon after diffusion from a nickel metal film deposited on the front surface of the wafer (T_{diff} = 1200°C, t = 30 min).

Studies of the effect of diffusion doping of monocrystalline silicon with nickel on the lifetime of the MCCs have shown (Table 1) that after additional thermal annealing τ in the control samples (group I) practically did not change (in the original samples, it was 5–7 μs). At the same time, in the samples in group II (nickel was deposited on the back-side of the SC) and the average value of τ after the diffusion of nickel increases by 1.35–1.4 times compared to the control samples. After thermal annealing τ increases by approximately another 20–25%. The total increase in τ is approximately 1.5–1.6 times relative to the control samples. In group III (nickel was sputtered on the front side), the average value of τ increases by 1.6–1.7 times after nickel diffusion (at T_{diff} = 1200°C). After additional thermal annealing, τ increases by an additional 30–35%; i.e., the total increase in τ is 1.9–2 times relative to the control SCs. It should be noted

Table 1. The lifetime of a nonconductive charge of all groups of SCs after nickel diffusion and thermal annealing and its change for groups II and III relative to the parameters of group I

Group	I	II	III
T_{diff} = 1200°C, t = 30 min	Annealing	Nickel diffusion	
τ, μs	5–6	7–8	9
$\Delta\tau/\tau_I$, times	–	1.35–1.4	1.6–1.7
T_{anneal} = 750–800°C, t = 1 hour	Additional thermal annealing		
τ, μs	5–7	9	12
$\Delta\tau/\tau_I$, times	1–1.15	1.5–1.55	1.9–2

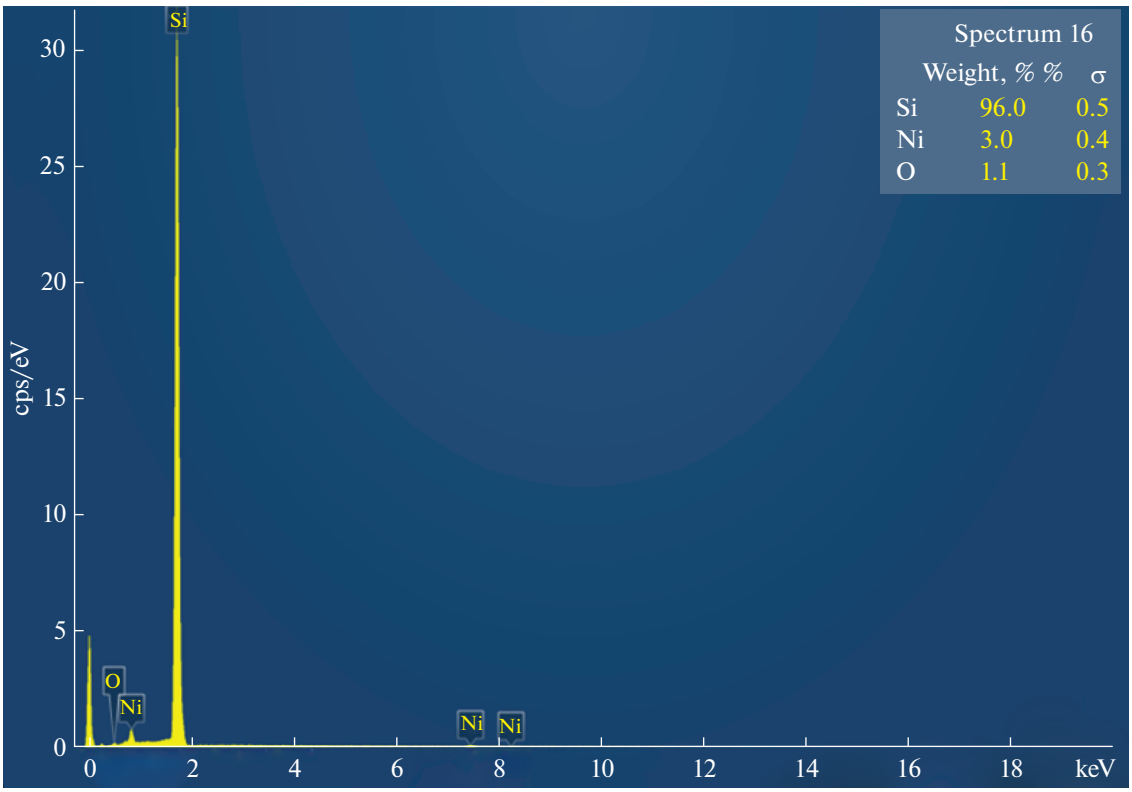


Fig. 2. Elemental composition of front surface of silicon samples diffusion-doped with nickel atoms

that the effect of increasing the lifetime of a nonconductive charge when doping an SC with nickel does not depend on the method of its introduction: before or after the creation of the *p–n*-transition. In Fig. 6 the VACs of SC groups I and III (nickel diffusion was carried out before the creation of the *p–n*-transition) are presented. It can be seen that the efficiency of an SC as a result of doping with nickel atoms increases by ~29% compared to the control sample (P_{max} increases from 12.08 mW/cm^2 up to 15.61 mW/cm^2, respectively). Experimentally, it has been established that the optimal temperature for nickel diffusion is $T_{diff} = 800–850$°C, and the temperature of additional thermal annealing is $T_{anneal} = 750–800$°C. The use of these technological modes makes it possible to increase the efficiency of silicon SCs by 25–30%, which is consistent with the results in [14, 15].

In order to establish the relative contribution of surface and bulk nickel clusters, the near-surface layer heavily alloyed with nickel, which was formed during its diffusion, was removed layer-by-layer. In these experiments, the *p–n*- transition was formed after removing the corresponding part of the enriched *Ni* layer. As can be seen from Fig. 7, with the increasing thickness of the removed facial layer Δx, the maximum removed power of the SCs in Group III decreases, and after removing a layer of 5 μm, the maximum power drops by ~20–25% compared to the same parameter for samples with $\Delta x = 0$ μm. The results of measuring the lifetime of the MCCs also showed that with an increase in the thickness of the removed front layer τ decreases monotonically, and after removing 1.5 μm, it remains practically unchanged. It should be noted that even after removing $\Delta x = 5$ μm, the value of τ in the SC data (24–26 μs) is 1.6–1.65 times greater than in the control SC (14–16 μs). The experimental results obtained indicate a significant decrease in the concentration of the recombination-active centers in SCs into which nickel impurities were diffused. This is apparently due to the gettering effect of both bulk and surface nickel clusters. Moreover, the presence of surface clusters in the samples makes a more significant contribution to increasing the efficiency of SCs compared to bulk clusters. In relation to this, we can conclude that the increase in the operational parameters of the SCs studied in this paper is largely due to the gettering effect of the surface layer with a nickel concentration of $n_S \sim 10^{20}–10^{21}$ cm^{-3} several micrometers thick, formed during the process of the diffusion of *Ni*.

As is well known [26], when creating an SC emitter by the diffusion of phosphorus, a dead layer is formed due to the formation of phosphorus silicides. This leads, on the one hand, to an increase in the resistance of the emitter layer, and on the other hand, to a decrease in the lifetime of the MCCs due to the generation of various types of structural defects in n^+-regions In relation to this, the presence of a nickel-enriched layer can also affect both the lifetime of the MCCs and the resis-

Elem.	Weight, %	σ, %	At, %
O	1.12	0.25	1.98
Si	95.89	0.45	96.58
Ni	2.99	0.38	1.44
Total:	100		100

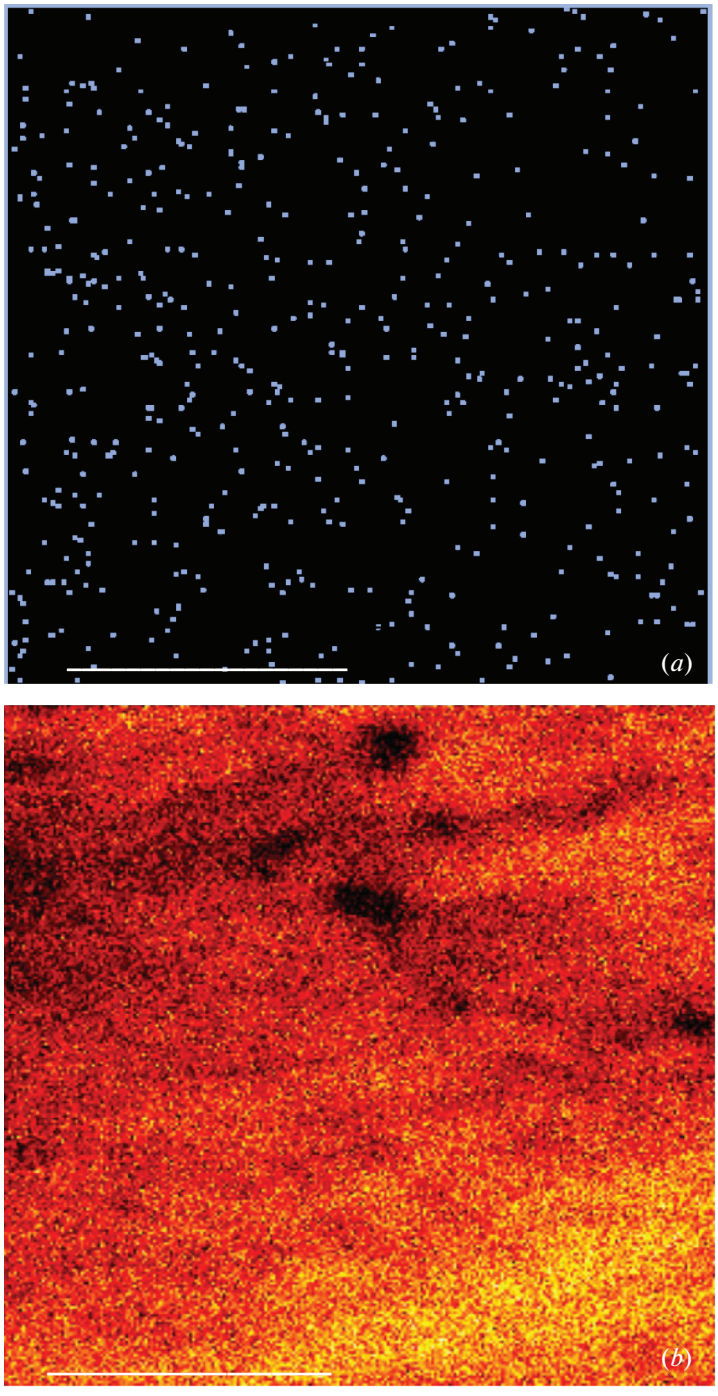


Fig. 3. Image of the surface of nickel-doped silicon obtained by the SIMS method for ions: (a) Ni^+ and (b) Si^+. (a) Bright dots correspond to clusters of nickel atoms; (b) bright dots correspond to silicon.

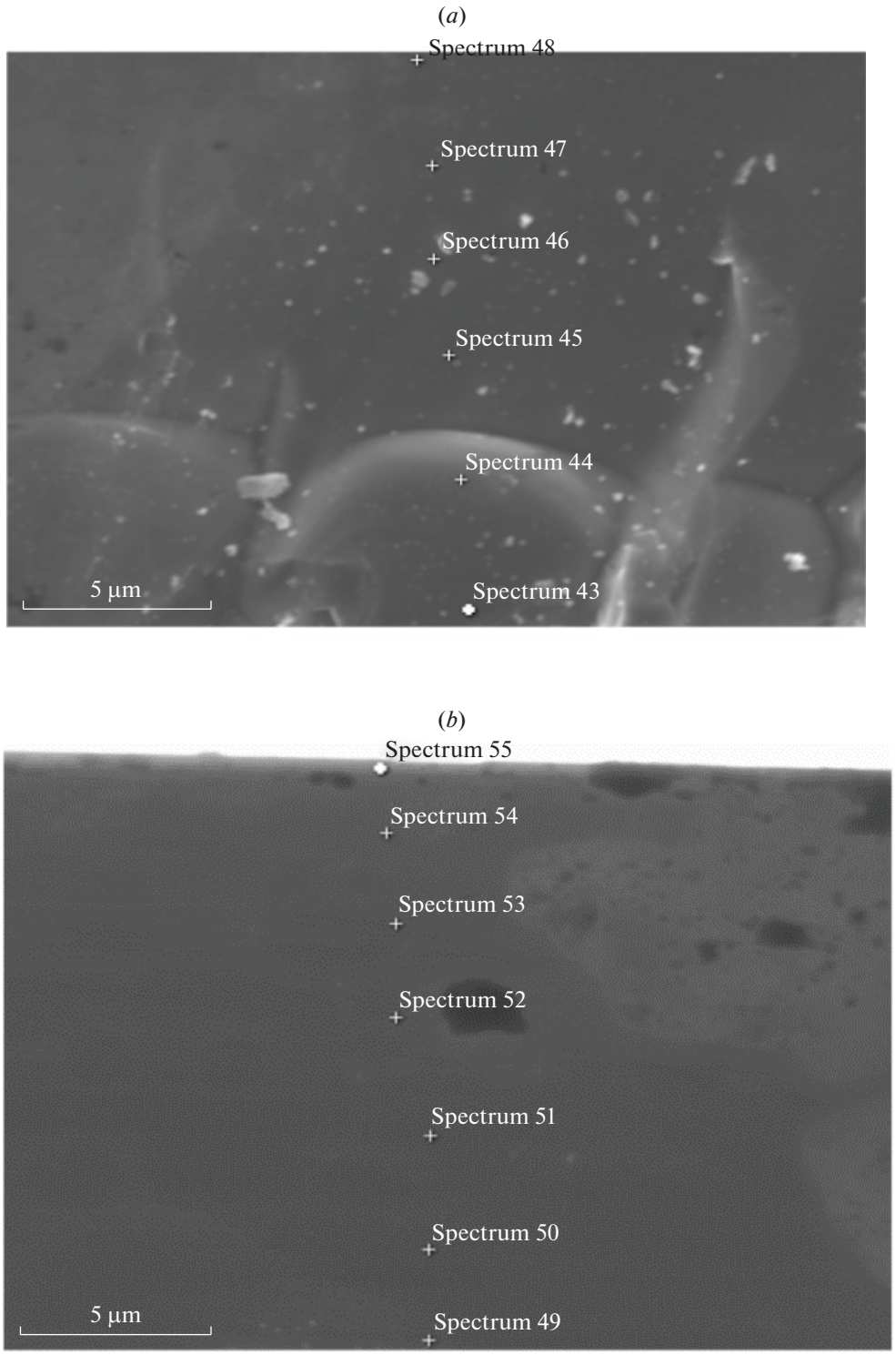
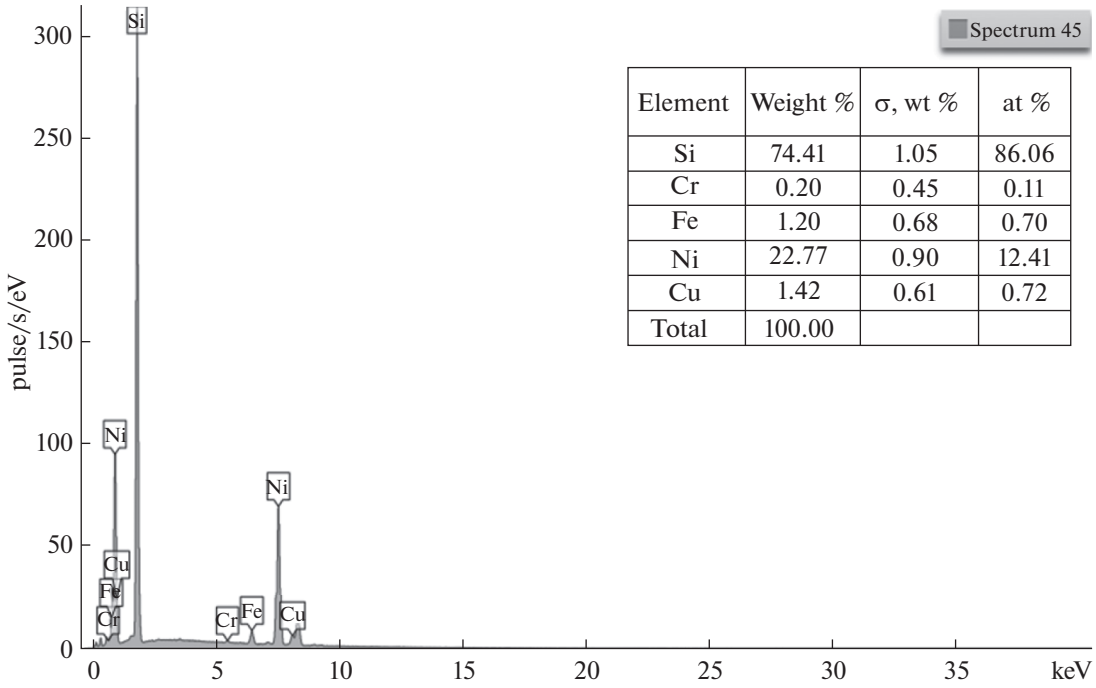


Fig. 4. Image of studied points of composition in silicon samples doped with nickel in the near-surface region (a) and volume (b) of the samples.

tance of the front layer of the SC. Due to the interaction of nickel with phosphorus in the near-surface layer, some of it will be excluded from the reactions of silicide formation. This will cause a decrease in the efficiency of recombination processes and an increase in the mobility of charge carriers in the emitter layer due to the elimination of the dead layer. A decrease in recombination leads to an increase in the concentration of optically generated charge carriers in the front layer of the SC and, as a consequence, to an increase



Element	Weight %	σ, wt %	at %
Si	74.41	1.05	86.06
Cr	0.20	0.45	0.11
Fe	1.20	0.68	0.70
Ni	22.77	0.90	12.41
Cu	1.42	0.61	0.72
Total	100.00		

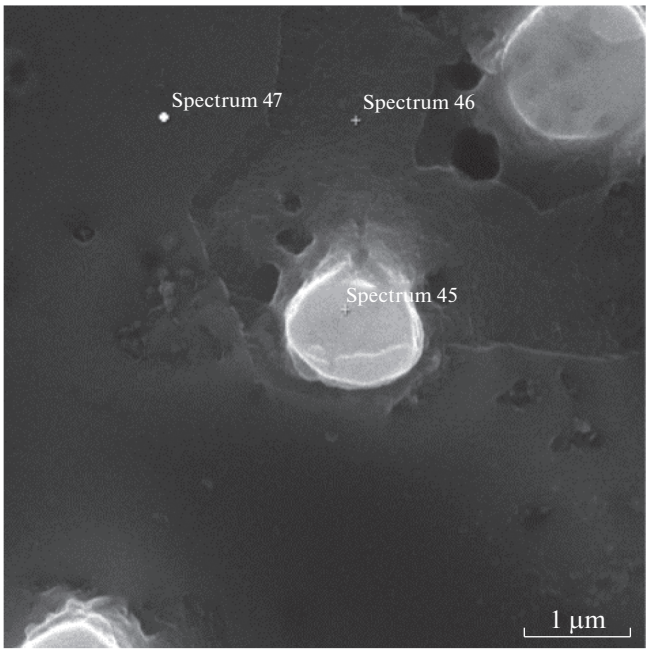
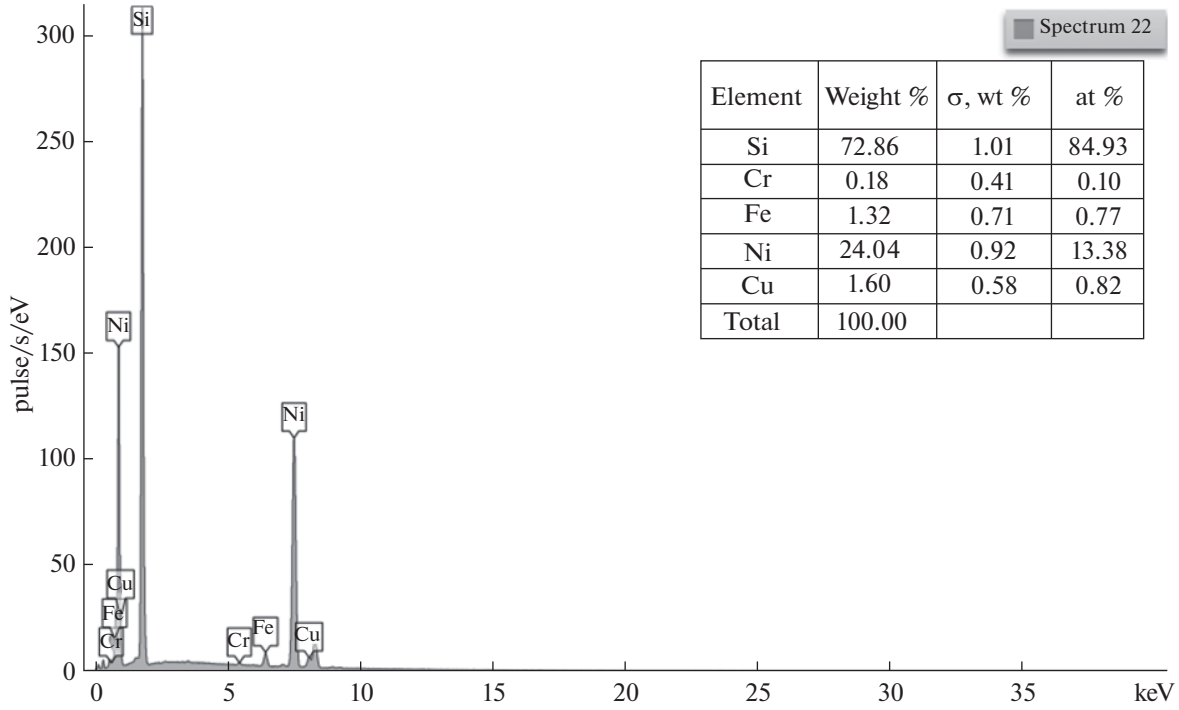


Fig. 5. Image and elemental composition of surface (a) and bulk (b) nickel clusters (obtained using X-ray local probe microanalysis) after additional thermal annealing at T_{anneal} = 800°C.

in the filling factor of the VAC (ξ). In addition, increasing the mobility in the emitter layer will reduce ohmic losses, which also increases ξ.

Measurement of the emitter surface resistance (n^+-layer) showed it decrease by 15–20% after the additional thermal annealing. In SCs having a nickel-enriched area on the front side of the p–n^+-transition, the filling factor of the VAC increased by 6–7% compared to the same characteristic for the control SC.

It should be noted that surface resistance measurements during the layer-by-layer removal of nickel-doped silicon (before the formation of the p–n transition) showed an insignificant effect of nickel on the volume resistance of silicon. This allows us to conclude that the majority of nickel atoms in the studied samples are not electroactive, which is consistent with the results of [9, 25]. The results obtained are due to the interaction of electrically neutral nickel impurity



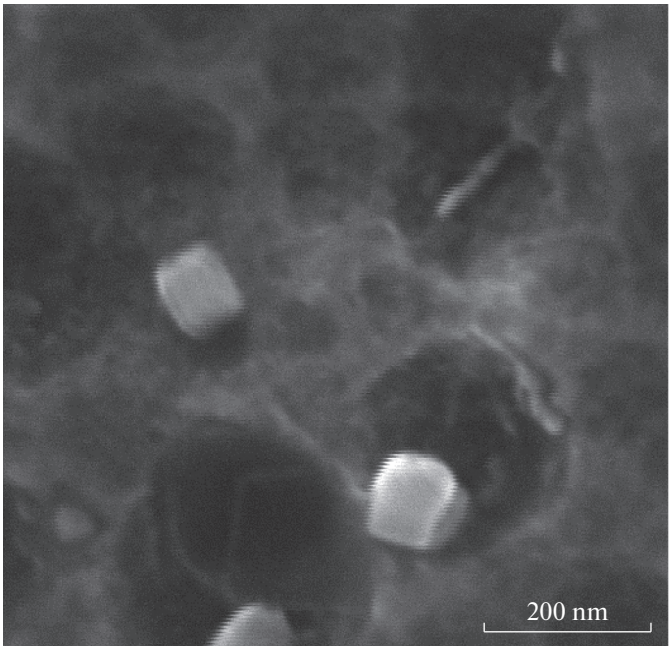
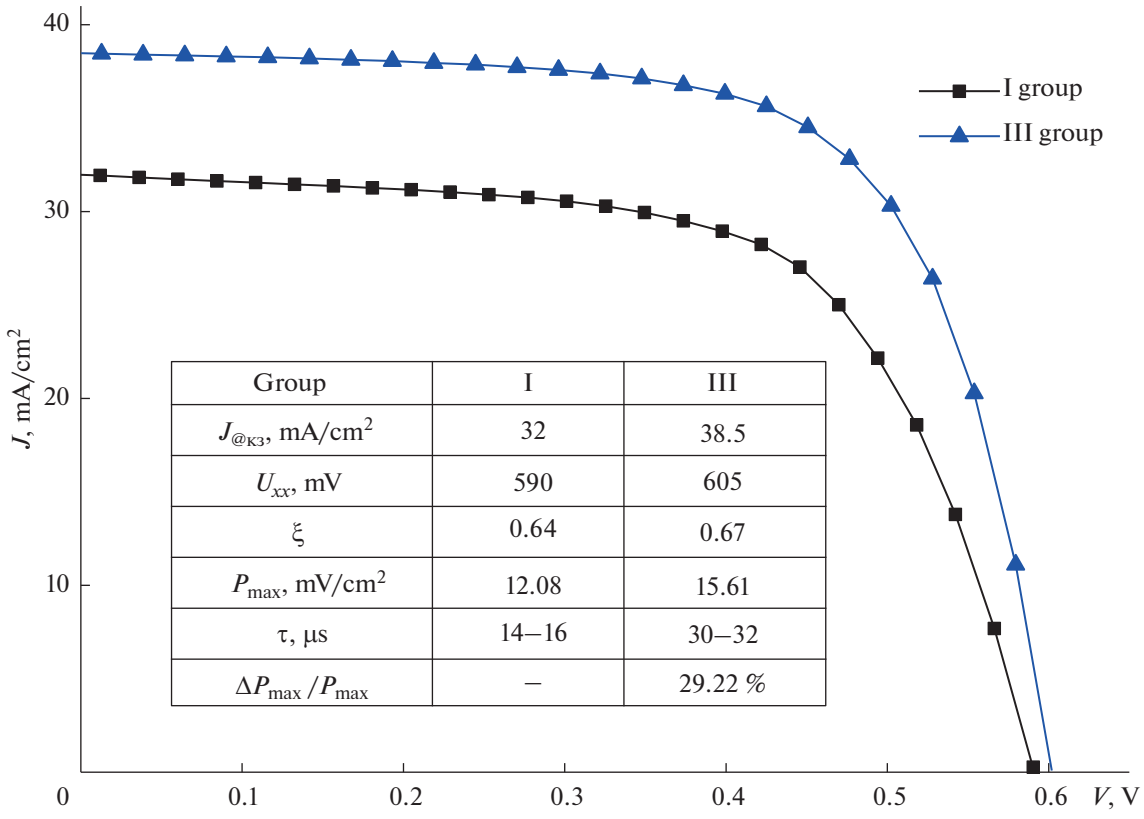


Fig. 5. (Contd.)

atoms located in the interstitial positions or clusters with technological impurities.

4. DISCUSSION OF EXPERIMENTAL RESULTS

As mentioned above, the nickel atoms introduced in the bulk of single-crystalline silicon by the diffusion method form clusters both in the near-surface region and in the bulk [22–25]. The centers of nucleation of nickel clusters are oxygen atoms and other silicon lattice defects, which are found in large numbers near the surface and also form in the diffusion layer of the n^+-SC type. These clusters act as getter centers for various uncontrolled impurities (O, Cu, Fe, Cr, Au [10, 15, 20]) and other defects of various kinds [25]. The main contribution to the energy of the interaction of impurity atoms with clusters of second-phase inclusions in semiconductors comes from elastic and elec-



Group	I	III
$J_{@КЗ}$, mA/cm^2	32	38.5
U_{xx}, mV	590	605
ξ	0.64	0.67
P_{max}, mV/cm^2	12.08	15.61
τ, μs	14–16	30–32
$\Delta P_{max}/P_{max}$	–	29.22 %

Fig. 6. VACs of solar and electrophysical parameters (table) of elements of Groups I and III after additional thermal annealing at T_{anneal} = 750–800 °C ($\Delta P_{max}/P_{max}$ is the relative change in maximum power compared to control samples).

trical interactions. The predominant interaction is elastic, caused by the difference in the atomic radii of the impurity and the matrix. Thus, the main driving force for the gettering effect of nickel clusters (and individual nickel atoms located in the interstitial positions) is the elastic stress fields that form around the clusters and are caused by the difference in the atomic radii of silicon and nickel (1.11 Å and 1.24 Å, respectively) [27]. It is energetically advantageous for atoms of technological impurities to coagulate on the dangling bonds around clusters, thus reducing the energy of the elastic deformation of the lattice. It should be noted that previously a similar effect of the gettering of technological impurities was observed when single-crystalline silicon was doped with rare-earth element (REE) impurities, which led, in particular, to an almost doubling of the energy resolution of ionizing radiation detectors made on monocrystalline silicon doped with REEs for α-particles of ^{239}Pu with an energy of 5.4 MeV due to the increase in the MCC lifetime in this material [28, 29].

The effect of gettering rapidly diffusing technological process impurities (TPIs) by nickel clusters is due to the following factors:

— there is always a large number of microdefects on the surface, which leads to the intense precipitation of nickel. Precipitates are practically pure metal and have an increased binding energy with TPIs (up to ~2.7 eV for single-crystalline nickel [30, 31]).

— in the near-surface region, the concentration of nickel atoms is higher than in the bulk by 2–3 orders of magnitude; therefore, the efficiency of gettering in the near-surface region is greater due to the larger number of formed clusters and precipitates [10–13];

— the gettering rate in the SC volume is lower due to the lower concentration of nickel clusters and the small number of precipitates. Considering the high diffusion coefficient of TPIs, they quickly reach the front and back surfaces of the SC, where there is a high concentration of nickel precipitates and clusters, as well as a higher binding energy of TPI atoms;

— the process of gettering by nickel clusters is enhanced by additional thermal annealing. Additional thermal annealing accelerates the achievement of equilibrium, but the very high annealing temperature (T_{anneal} > 900°C) leads to the disintegration of nickel clusters and precipitates [24–26].

In the process of the formation the *p–n* transition of the SC, as well as additional thermal annealing at T_{anneal} = 700–800°C, the nature of the distribution of nickel by volume remains virtually unchanged. From this we can conclude that the near-surface layer

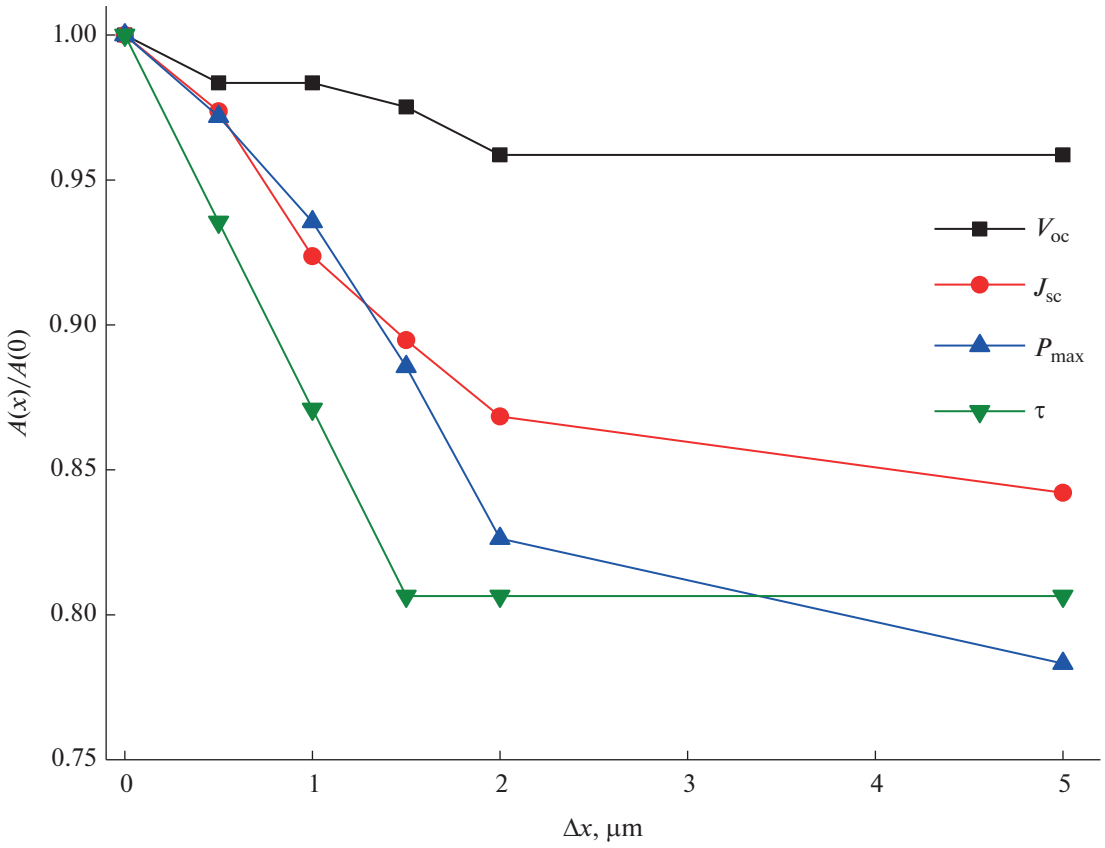


Fig. 7. Change $J_{\text{short circuit}}$, U_{xx}, $P_{\max}$ and τ SCs depending on the thickness of the removed near-surface nickel-enriched layer relative to the SC without removing the nickel-enriched layer.

enriched in nickel has a predominant influence on the parameters of the SC.

It is known [9, 22] that electrically neutral nickel atoms located in interstitial metastable states in the silicon lattice, both in the process of diffusion and during additional low-temperature thermal annealing, tend to a more energetically favorable state. During diffusion and cooling, electrically neutral nickel atoms form nuclei of clusters and precipitates, and additional thermal annealing activates the processes of formation and growth of cluster sizes. In the process of formation p–n transition of the SC, as well as additional thermal annealing at $T_{\text{anneal}} = 700$–At 800°C, the nature of the distribution of nickel by volume remains practically unchanged. From this we can conclude that the near-surface layer enriched in nickel has a predominant influence on the parameters of the SC.

It should be noted that the efficiency of reducing the concentration of recombination centers during gettering is directly related to the solubility limit of nickel atoms under technological diffusion conditions, since it is this value that determines the concentration of clusters. With the increasing diffusion temperature, the concentration of electrically neutral nickel atoms increases, the concentration of clusters increases, and, as a consequence, the concentration of recombination centers decreases [15]. However, as a rule, with the increasing temperature and duration of diffusion, the concentration of recombination centers also increases [7]. Consequently, the lifetime of the nonconductive charge in the SC decreases, and this leads to a decrease in efficiency. In addition, the formation of a p–n transition by the diffusion of phosphorus or boron is usually carried out at temperatures of 900–1050°C, which also always causes an increase in the concentration of recombination centers and a significant decrease in τ [1–4]. These factors determine the need to optimize the technological conditions for nickel gettering.

From a technological point of view, a layer of metallic nickel can also be applied to the silicon surface for gettering (on dozens of wafers simultaneously), and the diffusion of nickel atoms can be carried out in the open air at a relatively low temperature ($T_{\text{diff}} = 800$–850°C for 30 minutes). Based on this, the method of the preliminary doping of silicon with nickel from a chemically deposited layer may be promising for implementation in the industrial technology of SC production. This method, perhaps, without significant changes, can be combined with the standard technological processes for manufacturing SCs at modern electronics enterprises and will allow increas-

ing the efficiency of SCs based on monocrystalline silicon with little additional financial and material costs.

CONCLUSIONS

It has been established that doping the SCs of nickel atoms makes it possible to increase the lifetime of MCCs in the material by up to 2 times and the efficiency of solar energy conversion by 20–25%. The experimental results obtained are due to the interaction of electrically neutral nickel impurity atoms located in the interstitial positions or clusters with technological impurities. During the process of diffusion and subsequent precipitation annealing, nickel forms clusters both in the near-surface region and in the bulk of the samples. It is shown that the distribution of nickel clusters in the volume of the material is almost uniform, and their size does not exceed 0.5 μm. The concentration of clusters in the volume is $\sim 10^{11}$–10^{13} cm^{-3} and in the near-surface layer $\sim 10^{13}$–10^{15} cm^{-3}. It has been established that the decisive role in increasing the efficiency of SCs is played by the processes of gettering by nickel clusters of recombination-active rapidly diffusing technological impurities, occurring in the nickel-enriched front surface region of the SCs.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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