

Micro alloying of SiC by radioisotope

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Abstract. The endotaxia is the process of growth of one crystal structure inside the volume of another. In this case we are talking about the formation of the Silicon Carbide film in the Silicon substrate. The Silicon substrate is placed in the gas chamber. The sample is exposed to the stream of methane gas CH_4 at temperature of 1360 - 1380 °C and at normal pressure. Moreover, gas contains both the stable Carbon isotope C^{12} and the radioactive Carbon isotope C^{14} , and hydrogen H_2 in the gas acts as a carrier of Carbon.

1 Introduction

The development and research of low-power radioisotope power sources is a popular and relevant research area. These devices are necessary for powering autonomous systems, such as various remote sensors, beacons, remote stations, small spacecraft and nanosatellites, i.e. necessary where uninterrupted and low-power power is required for decades. There are various types of radioisotope power sources, differing in the principle of operation and design [1, 2]. The optimal design of the radiation-stimulated current source depends on the manufacturing technology of semiconductor structures. Among the technologies known, for example, the preparation of 3C-SiC by CVD [3, 4], where the gaseous substances are SiH_4 , C_3H_8 , SiH_2Cl_2 . In the above examples, the SiC film is epitaxially increased in the Si surface. Earlier in the works [5-9] in relation to the self-diffusion of C^{14} , it was experimentally shown that the diffusion intensity increases with doping. In this article, we consider the process of carbon diffusion in a silicon substrate, which is used in the manufacturing technology of the beta converter on the carbon radioisotope C^{14} [10], [11]. The uniqueness of this converter lies in the fact that the half-life of C^{14} radiocarbon is 5730 years. This means that an energy source working on the beta decay of this isotope will be able to power the necessary device for a time commensurate with human life. Therefore, the potential use of beta converters in medicine, namely, as power sources for pacemakers, deserves special attention.

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2 Carbon diffusion in silicon and solution of the Neumann problem

The endotaction process is the growth process of one crystalline structure within the volume of another. In this case, we are talking about the formation of a silicon carbide film on a silicon substrate. A silicon substrate is placed in the gas chamber. At a temperature of 1360 - 1380 °C and normal pressure, the sample is exposed to the flow of methane gas CH_4 . Moreover, the gas composition includes both the stable carbon isotope C^{12} and the radioactive carbon isotope C^{14} . The process lasts 1 hour, during which methane molecules react chemically with silicon atoms, forming silicon carbide and hydrogen



As a result, a silicon carbide SiC film grows on the surface of the sample, and the hydrogen formed during the reaction leaves the sample and is of no further interest [12]. As a result of a chemical reaction, the crystal lattice of the sample changes and, due to the different lattice parameters of Si and SiC, defects are formed [13].

Carbon atoms that have penetrated into the structure of the sample can undergo self-diffusion and diffuse deeply. A distinction is made between the interstitial diffusion mechanism, when an impurity atom moves from one interstitial site to an empty neighboring one, and the vacancy mechanism, when an atom moves from one lattice site to the site where the vacancy is located [14]. The diffusion mechanism in a solid is more dependent on the immediate environment of the impurity atom in the solvent. Distinguish between interstitial solutions and substitution solutions. In interstitial solutions, the impurity atom is predominantly located in the interstices of the lattice, although it can sometimes occupy the vacancy closest to itself. In substitutional solutions, the impurity atom, in most cases, occupies the lattice site before and after the jump.

We formulate the Neumann problem and consider its analytical solution for the diffusion process in a solid. In the experiment, at the initial time, the substrate does not contain carbon. Therefore, the initial condition will be such that the concentration of impurity atoms in the volume under consideration at the initial moment of time is zero. The boundary condition is set on the basis that in the process under consideration the flux density of the C^{12} impurity substance at the boundary is constant. As mentioned earlier, carbon atoms in reality can diffuse inside the substrate both by the vacancy mechanism and by the interstitial ones, but we are not aware of the ratio of these mechanisms. Therefore, speaking of the diffusion coefficient, we will talk about the effective diffusion coefficient, which includes both possible variants of diffusion in a solid. Thus, we set the task and we will formulate it in the following form:

$$\begin{aligned} \frac{\partial}{\partial t} c(x, t) &= D_{eff} \frac{\partial^2}{\partial x^2} c(x, t) \\ \frac{\partial c(0, t)}{\partial x} &= -\frac{J}{D_{eff}}, c(x, 0) = 0 \end{aligned} \quad (2)$$

where $c(x,t)$ is the concentration, D_{eff} is the effective diffusion coefficient independent of coordinates and temperature, x is the depth, J is the flux density of the substance at the boundary.

Analytical solving this problem with given additional conditions, we obtain a solution in the following form:

$$c(x, t) = J \frac{2e^{-\left(\frac{x^2}{4D_{eff}t}\right)} \sqrt{t}}{\sqrt{\pi D_{eff}}} - \frac{Jx}{D_{eff}} \operatorname{Erfc}\left(\frac{x}{2\sqrt{D_{eff}t}}\right) \quad (3)$$

where Erfc is the complementary error function that is obtained when solving the equation with this boundary condition.

3 SIMS results and calculation of model parameters

As experimental data we used one obtained by mass spectrometry of secondary ions of the surface of a silicon carbide film containing boron-doped carbon C^{12} . The investigated depth varies from $5.3 \cdot 10^{-8}$ to $24.3 \cdot 10^{-6}$ m. The concentration of atoms depending on the studied depth varies from $8.4 \cdot 10^{19}$ to $3.2 \cdot 10^{16}$ particles/cm³. The data are plotted and presented in Figure 1 [15].

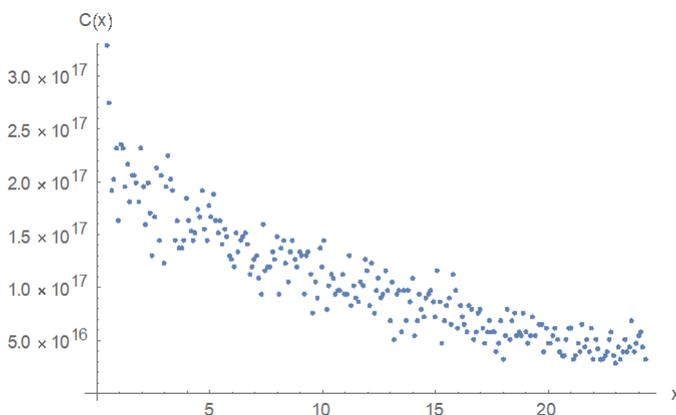


Figure 1. Experimental curve of the dependence of carbon concentration C^{12} on depth on the surface of p-SiC.

For calculation of model parameters we used the approximation method in the Wolfram Mathematica 11 software package. For a stable carbon isotope C^{12} , we get the impurity substance flux density at the sample boundary $J = 7.98532 \cdot 10^{14}$ particles/cm² · s and the effective diffusion coefficient for stable carbon $D_{eff} = 4.95984 \cdot 10^{-10}$ cm²/s. Values were obtained to solve the Neumann problem for a fixed $t = 60$ minutes.

Figure 2 shows three types of data: the experimental points according to the SIMS results are blue, the averaged curve of the experimental points is yellow, and the solution to the Neumann problem found with the parameters found by approximation is green.

4 Analysis and comparison of results

The resulting flux density J is actually the carbon flux density C^{12} at the substrate boundary. In reality, of course, a methane stream containing two carbon isotopes — C^{12} and C^{14} — falls onto the substrate boundary. But it follows from the physical meaning of the boundary condition that J is the flux density of the diffusing substance, which is considered when approximating the solution according to experimental data, i.e. carbon C^{12} . And for the numerical solution of the problem, this value was necessary to know. Therefore, this parameter was not fixed, but was selected by the approximation method. The value of the effective diffusion coefficient was also a parameter to be found. And the result obtained must be compared with known values. So, in the work of P. Werner, H.-J. Gossmann’s “Carbon diffusion in silicon” gives the values in the Arrhenius law for the carbon diffusion coefficient C^{12} separately

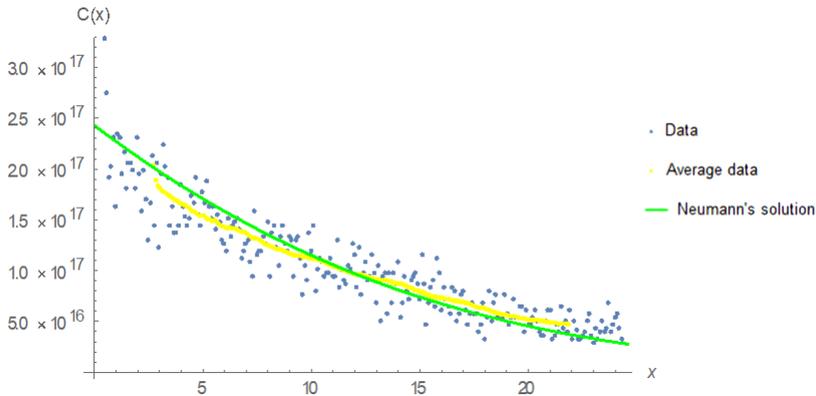


Figure 2. The result of approximation of the solution of the Neumann problem for given boundary and initial conditions in comparison with the average curve of the experimental points and with the initial experimental data. The accuracy of the approximations is at levels below 20%. The main part of the error is due to the relative spread of experimental points.

for the interstitial diffusion mechanism and the substitution mechanism. At a temperature value of $T = 1380\text{ }^{\circ}\text{C}$, the following values can be obtained [16]:

$$D_{int} = 9.79 \cdot 10^{-4} \text{ cm}^2/\text{s} - \text{diffusion coefficient for the interstitial diffusion mechanism;}$$

$$D_{vac} = 5.11 \cdot 10^{-10} \text{ cm}^2/\text{s} - \text{diffusion coefficient for the substitution mechanism.}$$

In this work, the found effective diffusion coefficient for carbon C^{12} is $4.95 \cdot 10^{-10} \text{ cm}^2/\text{s}$. From a comparison with the above data, it follows that the found effective carbon diffusion coefficient C^{12} is in good agreement with the value of carbon diffusion coefficient C^{12} for the vacancy diffusion mechanism in silicon. This means that under the given experimental conditions, during the diffusion process, the predominant mechanism is the substitution mechanism.

5 Summary

We conclude that at temperature $1360 \div 1380\text{ }^{\circ}\text{C}$ and normal pressure, annealing time $t = 60$ minutes, the C^{12} carbon flux density at the boundary $J = 7.98532 \cdot 10^{14} \text{ particles/cm}^2 \cdot \text{s}$ and the effective diffusion coefficient of carbon C^{12} $D_{eff} = 0.0495984 \mu\text{m}^2/\text{s}$ were obtained. The accuracy of the approximations is at levels below 20%. The main part of the error is due to the relative spread of experimental points. The results obtained are in good agreement with the known results.

Additional microalloying with radioisotope Carbon-14 at the level of ppm of Carbon-12 leads to new material properties influenced the subcritical value of concentration, which apparently corresponds to the transform (or absence) phase with Silicon and the effects of mobility and perhaps intensive self-diffusion of Carbon-14. The effects of beta radiation on the lower surface of silicon protected by a thin layer of photoresister confirm this. These properties allow to create effective heterostructures with different dark characteristics in relation to the light characteristics as in conventional photostructures.

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