St. Petersburg Polytechnic University Journal. Physics and Mathematics. 2023. Vol. 16. No. 1.1
Научно-технические ведомости СПбГПУ. Физико-математические науки. 16 (1.1) 2023

Conference materials
UDC 536.12
DOI: https://doi.org/10.18721/JPM.161.151

Numerical simulation of the temperature field distribution in the epitaxial graphene growth setup

S.P. Lebedev [1]✉, S.Iu. Priobrazhenskii [1], A.V. Plotnikov [1],
M.G. Mynbaeva [1], A.A. Lebedev [1]

[1] Ioffe Institute, St. Petersburg, Russia
✉ lebedev.sergey@mail.ioffe.ru

Abstract. An approach is presented to optimizing the growth of graphene on silicon carbide (SiC) substrates by using numerical simulation methods. The presented models in axisymmetric approximation show good convergence with experimental results and allow the studies of temperature fields inside closed growth cells. It is concluded that the use of numerical calculation methods is promising for optimizing the design of a technological setup for graphene growth by sublimation of the SiC surface.

Keywords: graphene, silicon carbide, simulation, temperature field distribution, sublimation growth

Funding: S.Iu. Priobrazhenskii acknowledge the support by the Russian Science Foundation grant 22-12-00134; S. P. Lebedev and A. A. Lebedev acknowledge the support by the Ministry of Science and Higher Education of the Russian Federation (agreement № 075-15-2021-1349).

Citation: Lebedev S.P., Priobrazhenskii S.Iu., Plotnikov A.V., Mynbaeva M.G., Lebedev A.A., Numerical simulation of the temperature field distribution in the epitaxial graphene growth setup, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 16 (1.1) (2023) 309–314. DOI: https://doi.org/10.18721/JPM.161.151

Материалы конференцииw
УДК 536.12
DOI: https://doi.org/10.18721/JPM.161.151

Численное моделирование распределения температурного поля в зоне роста графена, выращиваемого на SiC подложках

С.П. Лебедев [1]✉, С.Ю. Приображенский [1], А.В. Плотников [1],
М.Г. Мынбаева [1], А.А. Лебедев [1]

[1] Физико-технический институт им. А.Ф. Иоффе РАН, Санкт-Петербург, Россия
✉ lebedev.sergey@mail.ioffe.ru

Аннотация. В данной работе представлен подход к оптимизации роста графена на подложках карбида кремния (SiC) с использованием методов численного моделирования. Представленные модели в осесимметричном приближении показывают хорошую сходимость с экспериментальными результатами и позволяют проводить исследование температурных полей внутри закрытых ячеек. Сделан вывод о перспективности применения численных методов расчета для оптимизации конструкции технологической установки роста графена методом сублимации поверхности SiC.

Ключевые слова: графен, карбид кремния, моделирование, распределение температуры, сублимационный рост

Финансирование: С.Ю. Приображенский признателен за поддержку грантом РНФ 22-12-00134, Лебедев С.П. и Лебедев А.А. выражают благодарность Министерству науки и высшего образования Российской Федерации за поддержку (договор № 075-15-2021-1349).

Ссылка при цитировании: Лебедев С.П., Приображенский С.Ю., Плотников А.В., Мынбаева М.Г., Лебедев А.А. Численное моделирование распределения поля температур в установке эпитаксиального выращивания графена // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2023. Т. 16. № 1.1. С. 309–314. DOI: https://doi.org/10.18721/JPM.161.151

Introduction

One of the promising ways to obtain two-dimensional graphene films is the method of thermal decomposition (sublimation) of the surface of SiC substrates [1]. This method makes it possible to obtain high-quality graphene/SiC structures suitable for creation of various devices, for example, high-sensitivity sensor elements, whose principle of operation is based on change of resistance of graphene upon adsorption on its surface of low concentrations of bimolecular complexes and gaseous chemical compounds [2–4]. The creation technology of such devices imposes high requirements on the crystal perfection and uniformity of the thus produced graphene.

Growth of graphene layers by SiC surface sublimation flows in high-frequency (HF) induction heating systems at temperatures of 1500–2000 °C [5]. The temperature control in such technological setup is only possible on the surface of the closed growth cell with an optical pyrometer. At the same time, the distribution of the temperature field in the substrate area, which is not available for pyrometric measurements, plays a significant role on the quality and the uniformity of the grown graphene. It is especially important to control the temperature field while making larger the SiC substrates used.

One of the ways to solve this problem is to simulate the processes that take place in a growth setup with the help of specialized mathematical programs. For example, such approach has been successfully used to analyze and correct the growth of bulk crystals in closed systems [6–7]. The data obtained during the simulation describes a number of parameters, including the distribution of the temperature field in the growth area. Thus, this approach can be applied to the study of graphene growth conditions by sublimation of the SiC surface.

The aim of this study was to analyze the possibility of using numerical methods to calculate the thermal fields in the epitaxial graphene growth setup for the further optimization of the existing technology.

Construction of the model

The epitaxal graphene growth setup is a water-cooled quartz chamber with equipment made of graphite elements arranged inside (Fig. 1, *a*). The main element of the internal equipment, which is heated in the electromagnetic field of the inductor, is a heater made of dense grades of graphite. The heater is located inside the porous carbon material insulation. The insulation provides more uniform heating of the growth cell and protection of the external elements of the setup from overheating. The correctness of numerical simulation data depends on the accuracy of setting the parameters of materials and the geometric dimensions of all setup elements.

The COMSOL Multiphysics package was chosen as the working environment for calculating the growth cell heating conditions. A number of simplifications were made at the simulation which do not significantly change the results of calculation. An axisymmetric approximation was used to obtain the 3D structures by rotating the cross-cut of the model configuration [8–9]. Thus, the initial spiral configuration of the HF inductor was converted into a ring set. A number of internal boundaries were excluded for the internal equipment of the growth setup. These simplifications made it possible not only to reduce the dimension of the model by going over to simple 2D domains, but also to include additional physical modules and extend the parameters of solvers. This in turn resulted in an improved accuracy and speed of data acquisition, as well as reduced computing capacity requirements. Fig. 1, *b* shows an example of simulated 3D-image of the graphite equipment heated inside the growth setup.

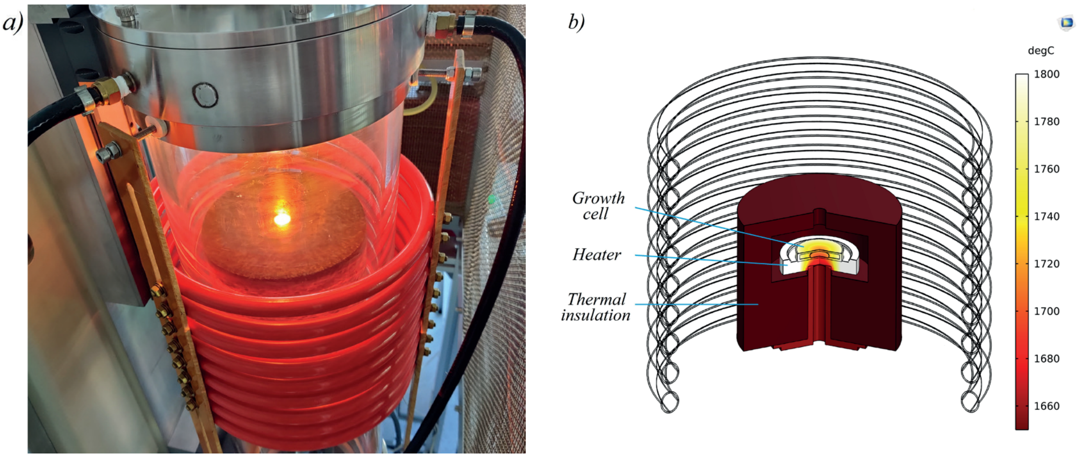

Fig. 1. Image of the epitaxal graphene growth setup (*a*), simulated 3D-image of the graphite equipment heated inside the growth setup (*b*)

Two configurations of the internal graphite equipment of the setup, which were applied in experiments to obtain epitaxial graphene, were used for calculations. In the first configuration, the heater is a graphite flat washer on which surface a growth cell with a substrate SiC is placed. In the second configuration, the heater has the shape of a tube, inside which a growth cell with a substrate is situated. Both heaters were made of graphite of the same grade and placed in thermal insulation made of fibrous graphite material.

Simulation results

To study the heating process, the temperature range from 1000 °C to 2000 °C was chosen, since all stages of graphene formation on the SiC surface occur in this temperature interval.

First stage of the study was to compare the simulation data with experimental heating data. The calculated temperatures were taken from the area of the growth cell, which is available for experimental pyrometric measurements. Experimental data was obtained using a Raytek Marathon MR1S infrared optical pyrometer operating in a two-color mode. The temperature measurement error does not exceed ±0.5% over the entire operating range. Fig. 2 shows heating and cooling graphs for two different equipment configurations. The simulation data show a fairly close similarity with the pyrometric data, which indicates that the model and the specification of the main characteristics of the materials used in the installation are correct. Small disarrangements between simulation and experimental data may be due to the imperfection of the design of real thermal insulation, as well as the use of some simplifications in the model, which were discussed above. Nevertheless, the convergence of the experimental and simulation curves and the coincidence of the key points allow further analysis of the thermal fields inside the growth setup.

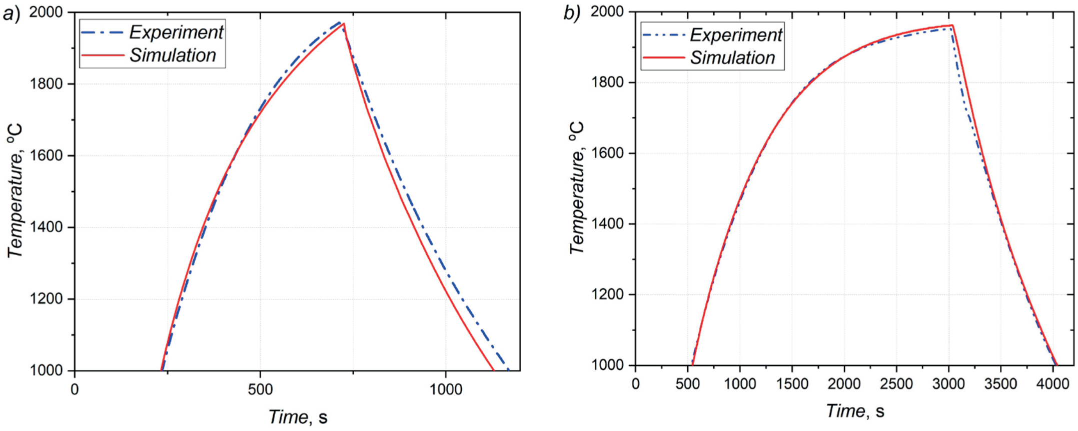

Fig. 2. Comparison of simulation and experimental heating curves for flat heater (*a*), tube heater (*b*)

The second stage of the study, the temperature distribution at the location of the SiC substrate in the growth cell for two heaters was compared. The growth cell is a flat graphite cup with a disk cover. The substrate is located in the cell in a special circular recess at the bottom, the diameter of which is 16 mm. The temperature value of 1700 °C in the central region of the substrate location was chosen as a general parameter for comparing the operation of two heaters, since this temperature is characterized by the onset of the formation of a graphene film on the SiC surface. Fig. 3 shows the temperature distribution of the cell cross-section heated by two heater configurations.

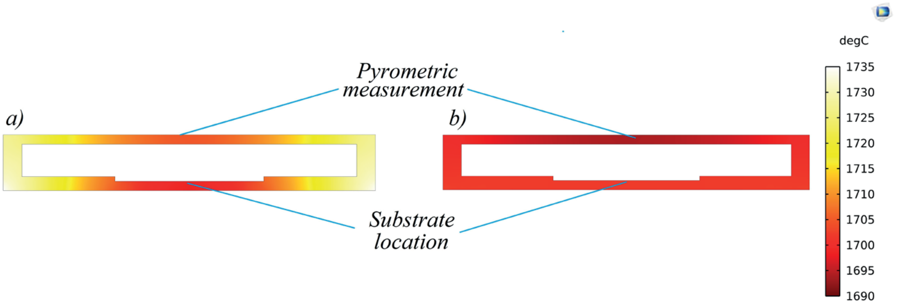

Fig. 3. Temperature distribution in growth cell heated in flat heater (*a*), tube heater (*b*)

Fig. 4, *a* shows images of the temperature distribution at the bottom of the growth cell obtained by simulation heating with different heaters. Fig. 4, *b*, for greater clarity, shows a graph of the temperature distribution in the substrate area. Based on these images, it can be concluded that the heating of the growth cell in the tube heater is much more uniform. At 1700 °C, the temperature differential in the substrate location for a tube heater is only 2–2.5 °C, and for a flat heater, this value increases to 15–16 °C. Thus, based on simulation data, it is possible to make a clear choice in favour of heating the cell in a tube heater.

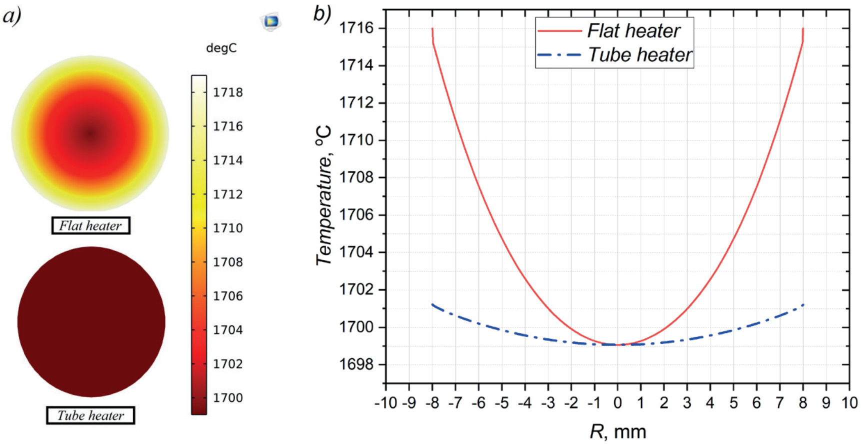

Fig. 4. The temperature distribution at the substrate location in the growth cell heated by flat and tube heaters (*a*), the temperature distribution graph by the cross-section of the bottom of the cell (*b*)

Another important heating parameter of the growth cell is the temperature difference between the area where the SiC substrate is situated (bottom of the growth cell) and the area where the temperature is measured with a pyrometer (growth cell cover). A large discrepancy between the temperatures in these regions may give no way of choosing the optimal mode for heating the SiC substrate to the graphene growth temperature, if being guided only by the pyrometer measurements. Simulation data make it possible to estimate the temperature difference and take it into account during the real growth process. Fig. 5 shows the temperature distribution for the substrate location and the growth cell cover. According to this graph, the temperature difference between the cover and the substrate during the growth of graphene should be about 7 °C.

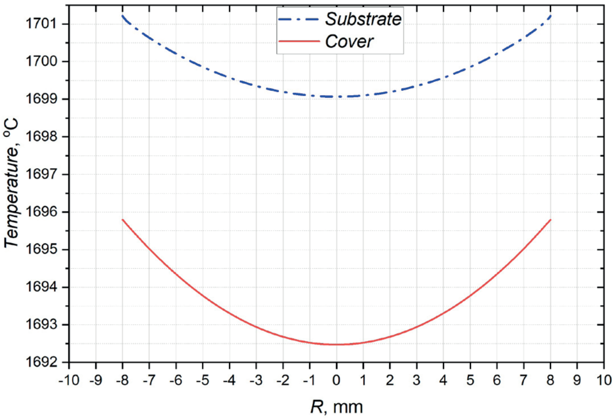

Fig. 5. Temperature distribution in different areas of the growth cell

Conclusion

The possibility of applying numerical methods for calculating thermal fields in the epitaxial graphene growth setup for the subsequent optimization of the existing technology is considered. A good convergence between the calculated and experimental curves of the growth cell surface temperature at the given generator power was demonstrated, which confirms that of the model construction and setting of the basic materials parameters of the setup are correct. Numerical simulation made it possible to compare different configurations of the internal graphene growth equipment, to determine a number of parameters of the thermal field in the growth cell for each case, and to identify the most optimal configuration.

The obtained results demonstrate the promise of using numerical simulation methods not only for determining the parameters of the thermal field in the growth setup, but also for optimizing the design of the setup for graphene growth by the SiC surface sublimation method. Through the use of the COMSOL Multiphysics package, it is possible to study many different options for the setup design and the features of its heating in a relatively short period of time. This factor may play a significant role in the development of this graphene growth technology and its transition from research to industrial production.

Acknowledgments

S. Iu. Priobrazhenskii acknowledge the support by the Russian Science Foundation grant 22-12-00134; S. P. Lebedev and A. A. Lebedev acknowledge the support by the Ministry of Science and Higher Education of the Russian Federation (agreement № 075-15-2021-1349).

REFERENCES

1. **Berger C., Song Z., LiT., Li X., Ogbazghi A.Y., Feng R., Dai Z., Marchenkov A.N., Conrad E.H., First P.N. and de Heer W.A.,** Ultrathin Epitaxial Graphite: 2D Electron Gas Properties and a Route toward Graphene-based Nanoelectronics, The Journal of Physical Chemistry B. 108 (52) (2004) 19912–19916.

2. **Liu Y., Dong X. and Chen Peng.,** Biological and chemical sensors based on graphene materials, Chemical Society Reviews. 41 (6) (2012) 2283—2307.

3. **Hill E.W., Vijayaragahvan A. and Novoselov K.,** Graphene Sensors, IEEE Sensors Journal. 11 (12) (2011) 3161–3170.

4. **Lebedev S.P., Usikov A., Novikov S., Shabunina E., Shmidt N.M., Barash I., Roenkov A.D., Lebedev A.A., Makarov Y.,** Graphene/SiC Functionalization for Blood Type Sensing Applications, Materials Science Forum. 924 (2018) 909–912.

5. **Lebedev A., Davydov V., Usachov D., Lebedev S., Smirnov A., Eliseyev I., Dunaevskiy M., Gushchina E., Bokai K., Pezold J.,** High Quality Graphene Grown by Sublimation on 4H-SiC (0001), Semiconductors. 52 (14) (2018) 1882–1885.

6. **Ramm M., Kulik A., Zhmakin I., Karpov S., Bord O., Demina S., Makarov Yu.,** Modeling of PVT growth of bulk SiC crystals: general trends and 2" to 4" reactor scaling, MRS Proceedings. 616 (2000) 227–233.

7. **Drachev R., Cherednichenko D., Khlebnikov I., Khlebnikov Y. and Sudarshan T.,** Graphitization of the Seeding Surface during the Heating Stage of SiC PVT Bulk Growth, Materials Science Forum. 433-436 (2003) 99–102.

8. **Narita T., Takahashi N., Horii M.,** Investigation of evolution strategy and optimization of induction heating model, IEEE Transactions on Magnetics. 36 (4) (2000) 1085–1088.

9. **Streblau M.,** Mathematical Model of Induction Heating Processes in Axial Symmetric Inductor-Detail Systems, TEM Journal. 3 (2) (2014) 162–166.

THE AUTHORS

LEBEDEV Sergey P.
lebedev.sergey@mail.ioffe.ru
ORCID: 0000-0002-5078-1322

PRIOBRAZHENSKII Sergei Iu.
sereyozha@yandex.ru

PLOTNIKOV Andrey V.
xdernx@gmail.com

MYNBAEVA Marina G.
mgm@mail.ioffe.ru
ORCID: 0000-0002-6321-1724

LEBEDEV Aleksander A.
shura.lebe@mail.ioffe.ru
ORCID: 0000-0003-0829-5053

Received 27.10.2022. Approved after reviewing 10.11.2022. Accepted 25.11.2022.