Conference materials
UDC 539.8:538.975
DOI: https://doi.org/10.18721/JPM.183.230

Long-term temperature resistance stability of gold-based multilayer electrodes for gas sensors

I.A. Lazdin[1✉], A.S. Kondrateva[2,1], I.M. Komarevtsev[2],
Ya.B. Enns[2], A.N. Kazakin[2], P. A. Karaseov[1]

[1] Peter the Great St. Petersburg Polytechnic University, St. Petersburg, Russia;

[2] Alferov University, St. Petersburg, Russia

✉ lazdin.ia@edu.spbstu.ru

Abstract. The long-term stability of resistive gas sensor microheaters was investigated.The microheaters were created as multilayered metallizations on a silicon oxide membrane. The behavior of Cr/Au/Cr and Cr/Ni/Au stacks was studied. The resistance of the Cr/Ni/Au stack stabilized after 15 hours of exposure due to gold burning into the chromium-nickel sublayer. Resistance drift is less than 7% after 48 hours of continuous operation at 350°C and 12% at 500°C. However, Cr/Au/Cr metallization cannot be used in high-temperature sensors because it degrades significantly at temperatures as low as 300°C. Stack capping with SiO_2 layer has low influence on initial forming stage and improves long-term stability.

Keywords: multilayer electrode, MEMS gas sensor, annealing, solid-state dewetting

Funding: Work was supported by Ministry of Science and Higher Education of Russian

Federation under the state assignment FSRM-2023-0009.

Citation: Lazdin I.A., Kondrateva A.S., Komarevtsev I.M., Enns Ya.B., Kazakin A.N., Karaseov P.A., Long-term temperature resistance stability of gold-based multilayer electrodes for gas sensors, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 18 (3.2) (2025) 155–158. DOI: https://doi.org/10.18721/JPM.183.230

Материалы конференции
УДК 539.8:538.975
DOI: https://doi.org/10.18721/JPM.183.230

Долговременная температурная стабильность многослойных электродов на основе золота для работы в газовых датчиках

И.А. Лаздин[1✉], А.С. Кондратьева[2,1], И.М. Комаревцев[2],
Я.Б. Эннс[2], А.Н. Казакин[2], П.А. Карасев[1]

[1] Санкт-Петербургский политехнический университет Петра Великого,

Санкт-Петербург, Россия;

[2] Академический университет им. Ж.И. Алфёрова РАН, Санкт-Петербург, Россия

✉ lazdin.ia@edu.spbstu.ru

Аннотация. Исследована долговременная стабильность микронагревателей резистивных газовых сенсоров. Микронагреватели создавались в виде многослойной металлизации на мембране оксида кремния. Изучено поведение стеков Cr/Au/Cr и Cr/Ni/Au как открытых, так и закрытых сверху слоем SiO_2. Сопротивление стека Cr/Ni/Au стабилизируется через 15 часов выдержки вследствие вжигания золота в хромоникелевый подслой. Дальнейший дрейф сопротивления при длительной эксплуатации при 350°С составляет менее 7%, при 500°C — 12% за 48 часов непрерывной работы. Металлизация Cr/Au/Cr не может быть использована в высокотемпературных сенсорах, поскольку существенно деградирует уже при 300°C. Покрытие нагревателя оксидом кремния не сказывается на его поведении на начальной стадии формования и улучшает долговременную стабильность.

Ключевые слова: многослойный электрод, газовый датчик, разрушение контактов

Финансирование: Работа выполнена в рамках Государственного задания «Активные композитные материалы и методики анализа для (био)сенсорики» (код темы FSRM-2023 0009).

Ссылка при цитировании: Лаздин И.А., Кондратьева А.С., Комаревцев И.М., Эннс Я.Б., Казакин А.Н., Карасев П.А. Долговременная температурная стабильность многослойных электродов на основе золота для работы в газовых датчиках // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2025. Т. 18. № 3.2. С. 155–158. DOI: https://doi.org/10.18721/JPM.183.230

Introduction

Change of a semiconducting film resistivity after adsorption of molecules of an analyte is widely used to produce resistive gas sensors. Operating temperature of such semiconducting active layer varies in the range 40 to 900 °C [1–4]. Therefore, high requirements are imposed on long term stability of heating elements. In order to minimize power consumption, thin membranes are used as a substrate of the sensing layer, which in turn is heated by micro-electrodes deposited directly on membrane [5]. At present, Pt is typically utilized as the heating layer material for high temperature applications. In order to minimize cost of a microheating element production, nickel, nichrome, gold and other materials are considered to substitute platinum. Noble metals are preferable, since they do not oxidize in air during operation. The main problem with thin coated electrodes is change in their parameters during high temperature exposure. Long-term instability of heating elements is caused mainly by partial or complete destruction of metal layer caused by solid state dewetting and diffusion. In this contribution, the influence of long-term annealing of gold-based microheaters on the resistance of these electrodes is investigated.

Materials and Methods

Microelectrodes were deposited onto the planar SiO_2/SiN membrane of a gas sensing chip by e-beam evaporation and photolithography. Two material stacks, namely, Cr-Au-Cr and Cr-Ni-Au, schematically drawn in the Fig. 1, *a* were formed. 5 nm thick chromium layer was deposited first in all cases. Gold layer in both cases was 100 nm thick, the nickel layer between gold and chromium was 20 nm thick, and the capping chromium layer was 5 nm thick. Fig. 1, *b* presents topology of electrodes. Some of heating tracks were subsequently covered by SiO_2 layer deposited by DC magnetron sputtering. Electric resistance of the as-grown electrodes varied from 300 to 500 Ohm.

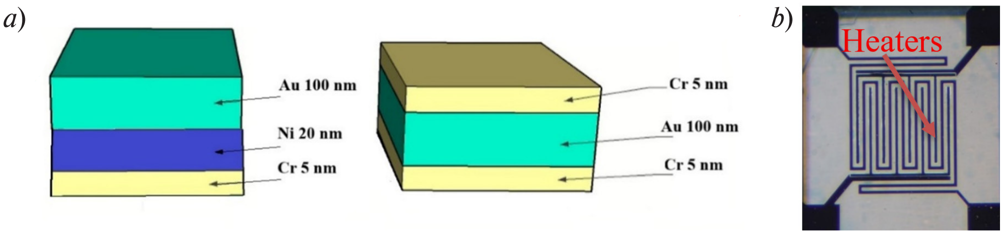

Fig 1. Stacks (*a*) and topology (*b*) of microheating electrodes

In order to investigate long term stability, artificial aging of the electrodes were used. Obtained structures were subjected to furnace thermal annealing in air ambient to assess the change in the heater resistance. The samples were loaded into furnaces preheated to required temperature and then abruptly removed from the hot furnace. Annealing temperatures were in the range from 300 to 500 °C. This is working temperature range of sensors detecting the most flammable and toxic compounds, so it is of interest to study the heater parameters at these temperatures.

Results and Discussion

Fig. 2 presents change in micro-heater resistances after annealing. It is seen from Fig. 2 that resistance of the heating elements start to grow up. Even at low annealing temperature, resistance of stack made of Cr/Au/Cr increases by an order of magnitude, which is more than others demonstrate (see Fig. 2). An increase in resistance is initially observed in the case of Cr/Ni/Au stack annealed at 350 °C (see Fig. 2, *b*). This process stops after 15 hours and only slight decrease in resistance is observed during further investigation. Increase of a heater resistance goes faster at 500 °C. Contrary to previous case, at this high temperature initial forming is followed by significant decrease in resistance (see Fig. 2, *c*), this process can be used to adjust the heating electrode parameters before use at lower temperatures.

Coating with 150 nm silicon oxide capping has no effect on initial resistance increase and improves heater stability during further annealing (compare Fig. 2, *b*, *d*). This capping prevents accessing the microheater by gases from atmosphere preventing possible oxidation of thin layers of Cr or Ni in the stack.

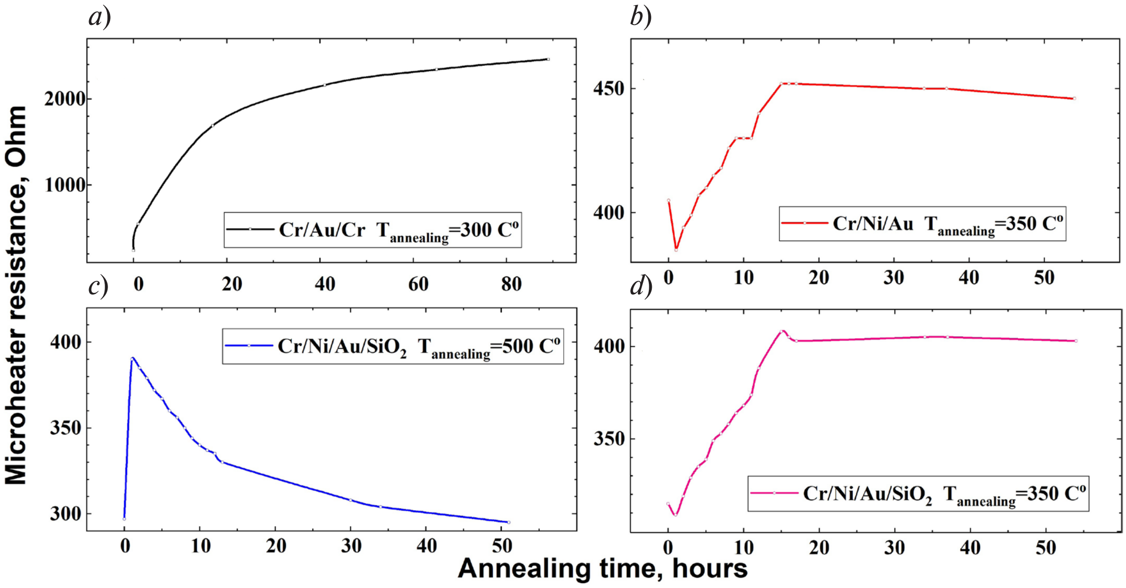

Fig. 2. Resistivity of electrodes annealed at different temperatures, as indicated

The experimental results demonstrate a significant effect of long-term annealing on the resistance of contact elements. According to previously published data, the main mechanism responsible for the change in resistance is dewetting – compression and partial destruction of a thin layer. It is widely observed for thin layers of materials with low adhesion to the surface at temperatures well below the melting point of the metal. Dewetting mechanisms for single-layer contacts made of various materials with high chemical stability (Au, Pt, Ag) are presented in detail in [6]. In our case of Cr/Au/Cr and Cr/Ni/Au stacks, same effect could appear leading to a decrease in the cross-section of the conductive layer and as a consequence to an increase in resistance. It is demonstrated that identical results of changing contact parameters.

When the annealing temperature of Cr/Ni/Au/SiO_2 stack is increased to 500 °C (see Fig. 2, ***c***) an initial sharp increase in resistance is observed due to dewetting, and a subsequent long-term decrease due to the slow process of interdiffusion of the Ni sublayer into Au. For a stack without an insulating layer of silicon dioxide, the trend of resistance change over time at a temperature of 500 °C is similar to the graph in Fig. 2, *c*. Conductive layer resistivity returns to its original value as a result of overgrowing along the interface of the two metals, with the formation of a Ni-Au solid solution similar to reported in [7].

Thus, the data obtained above show the need for a long-term temperature procedure for micro-heating elements based on the presented stacks for their stable operation at elevated temperatures.

Conclusion

Performance of Cr/Au/Cr and Cr/Ni/Au structures used as heating microelectrodes for gas sensor and other microelectronic devices operating at temperatures ranging from 40 to 500 °C was evaluated. The least suitable stack option for micro-heating elements is configuration Cr/Au/Cr. Thin (~5 nm) chromium layer does not prevent the diffusion of gold into the substrate and the disintegration of gold conductive layer during prolonged heating above 300 °C. The Cr/Ni/Au layer is a good choice for applications operating at temperatures up to 350 °C. Aging at 500 °C could be used for the resistance tuning. The obtained time characteristics can also be used to recalculate the aging time of the sample at lower operating temperatures.

Acknowledgments

Work was supported by Ministry of Science and Higher Education of Russian Federation under the state assignment FSRM-2023-0009.

REFERENCES

1. **Liu W., Gu D., Zhang J., Li X., et al.,** ZnSe/NiO heterostructure-based chemiresistive-type sensors for low-concentration NO_2 detection, Rare Metals. 40 (2021) 1632–1641.
2. **Srivastava S., Gangwar A.K., Kumar A., et al.,** Room temperature RF magnetron sputtered nanocrystalline NiO thin films for highly responsive and selective H_2S gas sensing at low ppm concentrations, Materials Research Bulletin. 165 (2023) 112330.
3. **Vorobyeva N., Rumyantseva M., Platonov V., et al.,** Ga_2O_3 (Sn) oxides for high-temperature gas sensors, Nanomaterials. 11 (11) (2021) 2938.
4. **Leech P.W., Kibel M.H., Barlow A.J., et al.,** Ohmic contacts to *n*-type 3C-SiC using Cr/Ni/Au and Ni/Cr/Au metallizations, Microelectronic Engineering. 215 (2019) 111016.
5. **Komarevtsev I., Kondrateva A., Lazdin I.A., et al.,** Microelectromechanical gas sensor of resistive type for detection of hydrogen sulfide down to low concentrations, Materials Physics and Mechanics. 52 (4) (2024) 9–22.
6. **Greenbank W., Hirsch L., Chambon S.,** Electrode de-wetting as a failure mechanism in thermally-aged OPV devices, Solar Energy Materials and Solar Cells. 178 (2018) 8–14.
7. **Bogatyrenko S.,** Formation of the solid solutions in the Au-Ni film system: In situ TEM study, Technical Physics. 59 (9) (2014) 1374–1377.

THE AUTHORS

LAZDIN Ilya A.
lazdin.ia@edu.spbstu.ru
ORCID: 0009-0002-0656-8433

KONDRATEVA Anastasia S.
kondrateva_n@spbau.ru
ORCID: 0000-0003-3915-9329

KOMAREVTSEV Ivan M.
imk@spbstu.ru
ORCID: 0000-0001-5118-8152

ENNS Yakov B.
Ennsjb@gmail.ru
ORCID: 0000-0003-4396-2368

KAZAKIN Alexei N.
Kazakin75@gmail.ru
ORCID: 0000-0001-8762-1587

KARASEOV Platon A.
platon.karaseov@spbstu.ru
ORCID: 0000-0003-2511-0188

Received 11.08.2025. Approved after reviewing 25.09.2025. Accepted 25.09.2025.