

Highly Stable Pt-activated Graphene Gas Sensors with Antimony Oxide Coatings

Ahmet Burak Baloglu, Margus Kodu, Tauno Kahro, Rainer Pärna, Aarne Kasikov, Raivo Jaanisoo
University of Tartu, Ülikooli 18, Tartu, Estonia
ahmet.burak.baloglu@ut.ee

Summary:

We report a significant improvement in the thermal stability of graphene-based gas sensors. By depositing a thin antimony oxide layer on graphene via pulsed laser deposition, a protective effect was observed at operating temperatures above 200 °C under humid air and harsh ozone conditions over extended periods, enabling sensors to maintain a stable response. Importantly, this protective layer preserves the CO-sensing capability introduced by Pt functionalization, indicating that the typical sensitivity-stability trade-off can be effectively mitigated without compromising the response to target gases.

Keywords: graphene, antimony oxide, thermal stability, CO detection, chemiresistor

Background, Motivation and Objective

Graphene has emerged as a promising material for next-generation gas sensors, due to its exceptional electronic characteristics and atomically thin structure. However, in its pristine state, graphene is relatively inert, which restricts its effectiveness as a receptor in chemiresistive gas sensors. Previous studies have demonstrated that introducing a functional layer, such as metals [1] or metal oxides [2], onto the graphene can substantially enhance its sensitivity to various toxic and pollutant gases by providing active receptor sites. Despite these improvements, such functionalization often introduces a trade-off between sensitivity and stability, as surface modifications can render sensors susceptible to degradation, particularly during long-term operation at elevated temperatures.

Thermal activation is a widely employed strategy to enhance the performance of both traditional metal-oxide and graphene-based gas sensors [3]. By activating the receptor layer, heating can significantly improve sensitivity and accelerate response and recovery dynamics. Furthermore, temperature modulation strategies have been extensively investigated to enhance selectivity and are already implemented in commercial metal-oxide-based sensing platforms [4]. However, prolonged exposure to elevated temperatures is known to reduce the operational lifetime of the chemiresistive gas sensors. Therefore, ensuring reliable long-term performance and extending the upper temperature limits requires effective protection of the active sensing layer.

In this study, we aimed to improve the long-term stability of graphene-based gas sensors for high-

temperature operations in humid air and harsh ozone environments, which we believe is a critical step toward reliable operation as commercial gas sensors.

Description of the New Method or System

Here, we report, for the first time, the protective role of thin antimony oxide layers deposited on graphene-based gas sensors via pulsed laser deposition (PLD) (Figure 1).

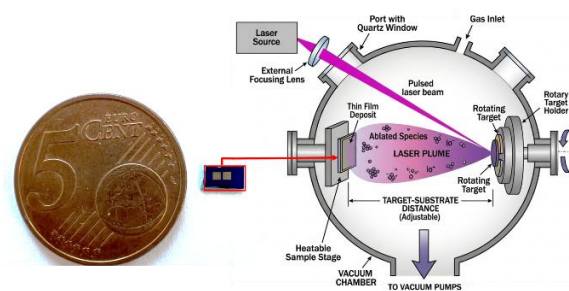


Fig. 1. PLD of graphene sensor. Graphene was previously wet-transferred between the Au electrodes on a Si/SiO₂ substrate (outlined in red). A 5-cent coin for scale.

Our results show that graphene sensors exhibit sensitivity to CO gas only after functionalization with Pt; however, significant degradation occurs at operating temperatures above 200 °C. In contrast, incorporating the antimony oxide interlayer enables the sensor to maintain performance at elevated temperatures without compromising CO sensitivity. This protective effect is highly reproducible, and the improved stability persists for at least two years, indicating that antimony oxide is a promising material for overcoming the long-standing stability limitations in gas sensor research.

Results

Figure 2 demonstrates the change in “relative response,” which is the variation of the electrical conductance in time, upon target gas (or heating), and is defined as:

$$\text{Relative response (\%)} = \frac{I_{\text{gas}} - I_{\text{air}}}{I_{\text{air}}} \times 100 \quad (1)$$

where I_{gas} is the conductance upon a target gas, while the I_{air} is the initial conductance in synthetic air.

Deposition of the Pt layer via PLD enabled the graphene sensor to detect CO gas even at room temperature (RT).

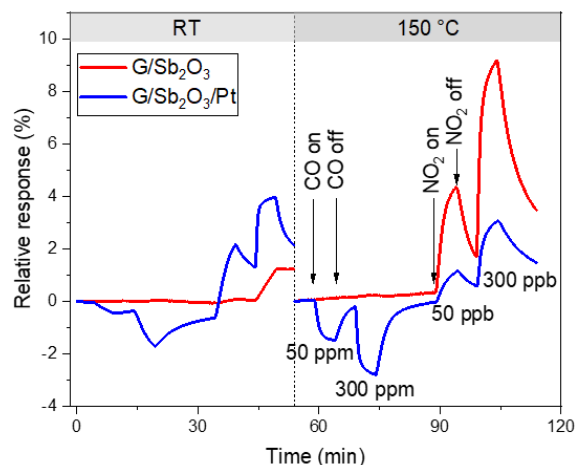


Fig. 2. Relative responses to CO and NO₂ gases in different concentrations at RT and 150 °C.

The long-term stability of the graphene sensors with and without the protective antimony oxide layer is shown in Figure 3. At 200 °C, the unprotected sensor current begins to drop, and at 250 °C, a significant and irreversible decrease occurs, indicating sensor degradation.

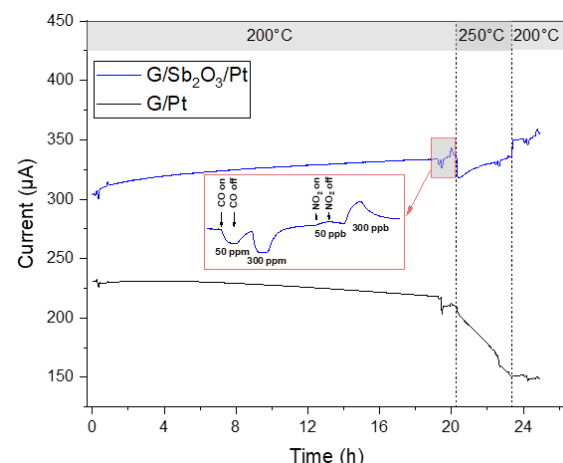


Fig. 3. Long-term stability of the graphene sensors in 40% humid air above 200 °C.

The influence of a harsh environment was investigated by elevating the temperature under an ozone atmosphere. Figure 4 shows that the antimony oxide-coated sensor achieved a stable baseline up to 200 °C, whereas the uncoated sensor exhibited a dramatic degradation trend.

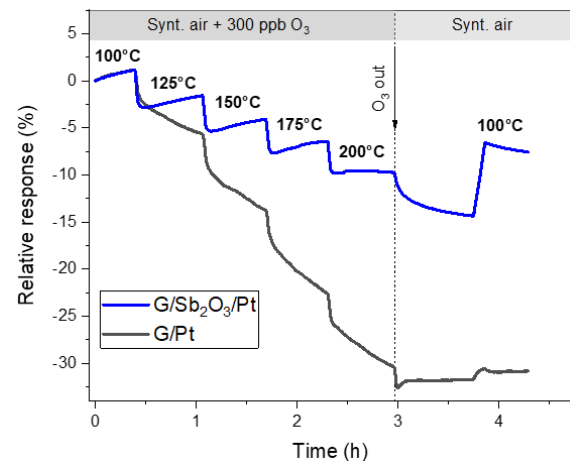


Fig. 4. Stability of the sensors (w/ and wo/ antimony oxide) under an ozone environment.

References

- [1] Kodu M, Berholts A, Kahro T, Avarmaa T, Kasikov A, Niilisk A, Alles H, Jaaniso R., Highly sensitive NO₂ sensors by pulsed laser deposition on graphene, *Applied Physics Letters* 12;109(11) (2016); doi: 10.1063/1.4962959
- [2] Berholts, A., Kodu, M., Rubin, P., Kahro, T., Alles, H. and Jaaniso, R., Layered heterostructure of graphene and TiO₂ as a highly sensitive and stable photoassisted NO₂ sensor, *ACS Applied Materials & Interfaces*, 16(33), pp.43827-43837 (2024); doi: 10.1021/acsami.4c08151
- [3] Vafaei, P., Lind, M., Kiisk, V., Kahro, T., Baloglu, A.B., Kodu, M., Raabe, R., Kasikov, A., Kikas, A., Kozlova, J. and Jaaniso, R., High-Performance Graphene-Based Gas Sensors with Pulsed Heating and AI Processing. *Advanced Sensor Research*, 4(12), p.e00083 (2025); doi: 10.1002/adsr.202500083
- [4] BME690 gas sensor (BST-BME690-DS001) datasheet, *Bosch Sensortec* (2025); URL: <https://www.bosch-sensortec.com/en/products/environmental-sensors/gas-sensors/bme690>

Acknowledgements

This research was funded by the Estonian Research Council grant (PRG1580).