Electrical Conductivity Modeling of SiO_2/CuBr Composite Sensitive Layer for Ammonia Gas Sensor Application

Caroline LAMBERT-MAURIAT[1],
Princesse N'Nang M'Boukou, Lisa Weber, Virginie Martini, Marc Bendahan

[1]Aix Marseille Univ, Université de Toulon, CNRS, IM2NP
Marseille (France)

E-mail contact: **caroline.mauriat@im2np.fr**

IARIA

Institut Matériaux Microélectronique Nanosciences de Provence

Caroline Lambert-Mauriat

Caroline Lambert-Mauriat is Associate Professor at IM2NP since 2000. She specializes in modeling and numerical simulation.

Her research focuses on understanding adsorption mechanisms, chemical reactivity, conduction in composites material and interfacial phenomena.

Her work is part of the theme of developing gas sensor applications within the ‘Microsensors & Instrumentation’ team at the IM2NP laboratory (UMR CNRS 7334).

I. Research Issue

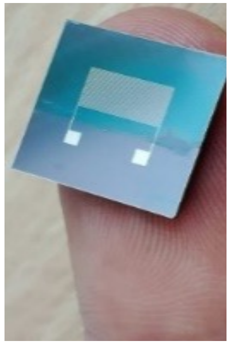
Detection of skin-emitted ammonia (NH_3) on patients suffering from chronic kidney disease could be an alternative to urine or blood tests

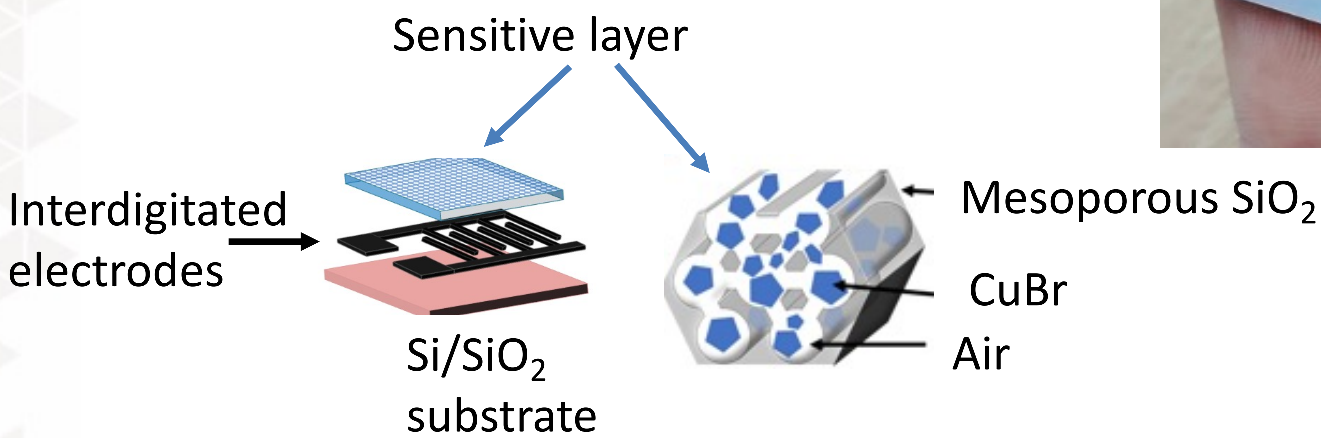
+

CuBr-based sensors are very selective to NH_3 at room temperature:

$Cu^{+} + NH_3 \rightarrow [Cu(NH_3)_2]^{+}$ complex

Microsensor development [1]



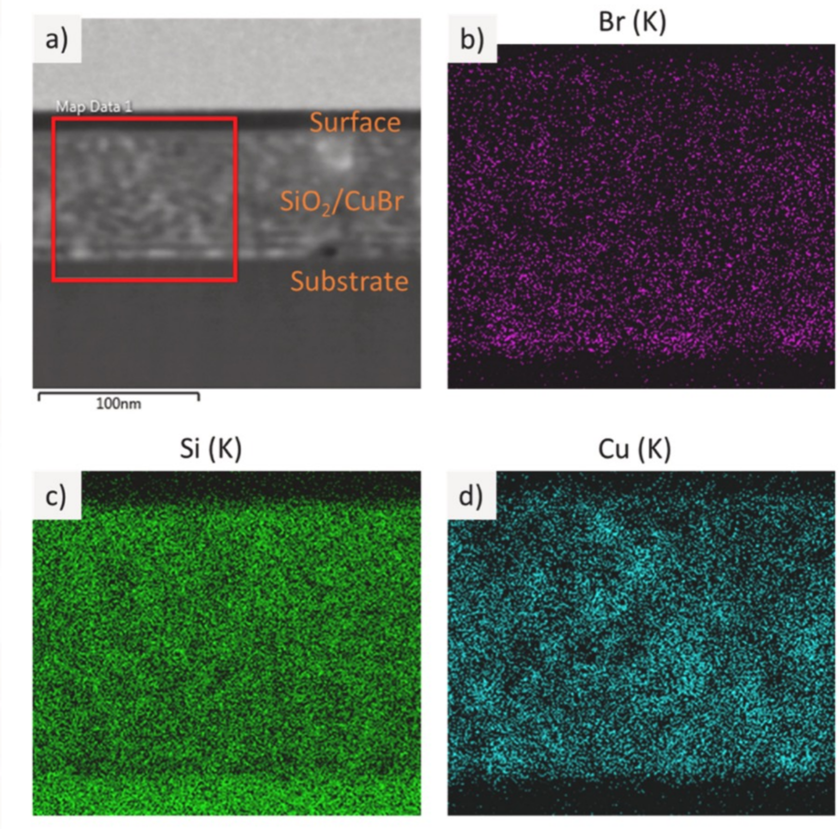


The objective of this work is to modeling electrical conduction within the sensitive layer

[1] L. Weber and *al.*, "A new approach for selective and ultrasensitive ammonia sensors by CuBr encapsulation in a mesoporous thin film for potential metabolic acidosis non-invasive monitoring", Sensors and Actuators B (vol. 417), pp 136124 (2024)

II.A. Numerical Model and Methods: Mesoporous Layer Modeling

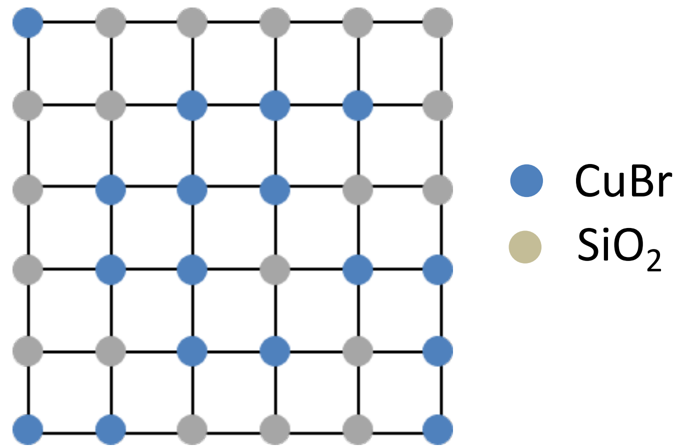
EDX mapping of the CuBr-impregnated SiO_2 mesoporous layer: distribution of the K line, for Br (b), Si (c) and Cu (d) atoms, in the area outlined in red on the image obtained by STEM (a), from ref. [1].



CuBr is uniformly distributed across the entire thickness

1st approximation

Cubic network of sites randomly occupied by SiO_2 or CuBr is considered



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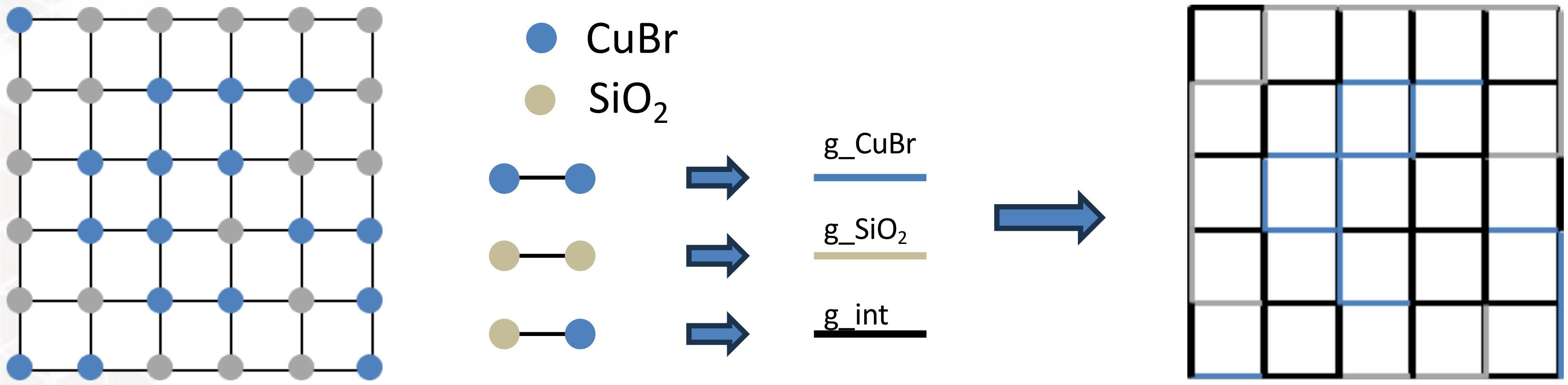
II.B. Numerical Model and Methods: Conductance Network

Cubic network of sites

Conductance Network



Values of conductances

Experiments

10^{-3} S.m^{-1} < g_CuBr < 10 S.m^{-1}

10^{-16} S.m^{-1} < g_SiO_2 < 10^{-12} S.m^{-1}

g_int is unknown

Used in our calculations

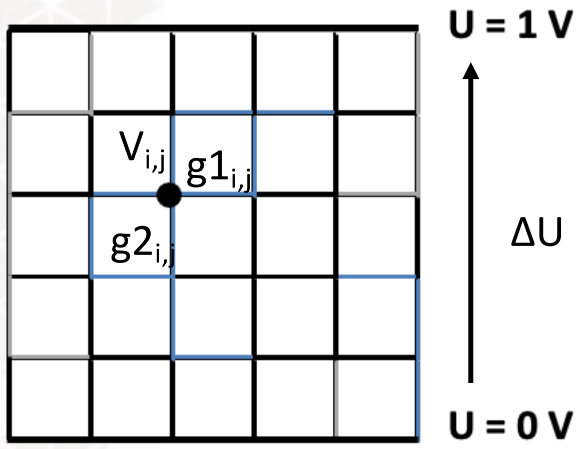
g_CuBr = 1 S.m^{-1}

g_SiO_2 = 1.10^{-9} S.m^{-1}

g_por = g_air = 1.10^{-12} S.m^{-1}

g_int = 10 S.m^{-1}

II.B. Numerical Model and Methods: Total Conductivity Calculation



Limit conditions

- Polarisation voltage ΔU = 1 V
- Boundary conditions in perpendicular direction to ΔU

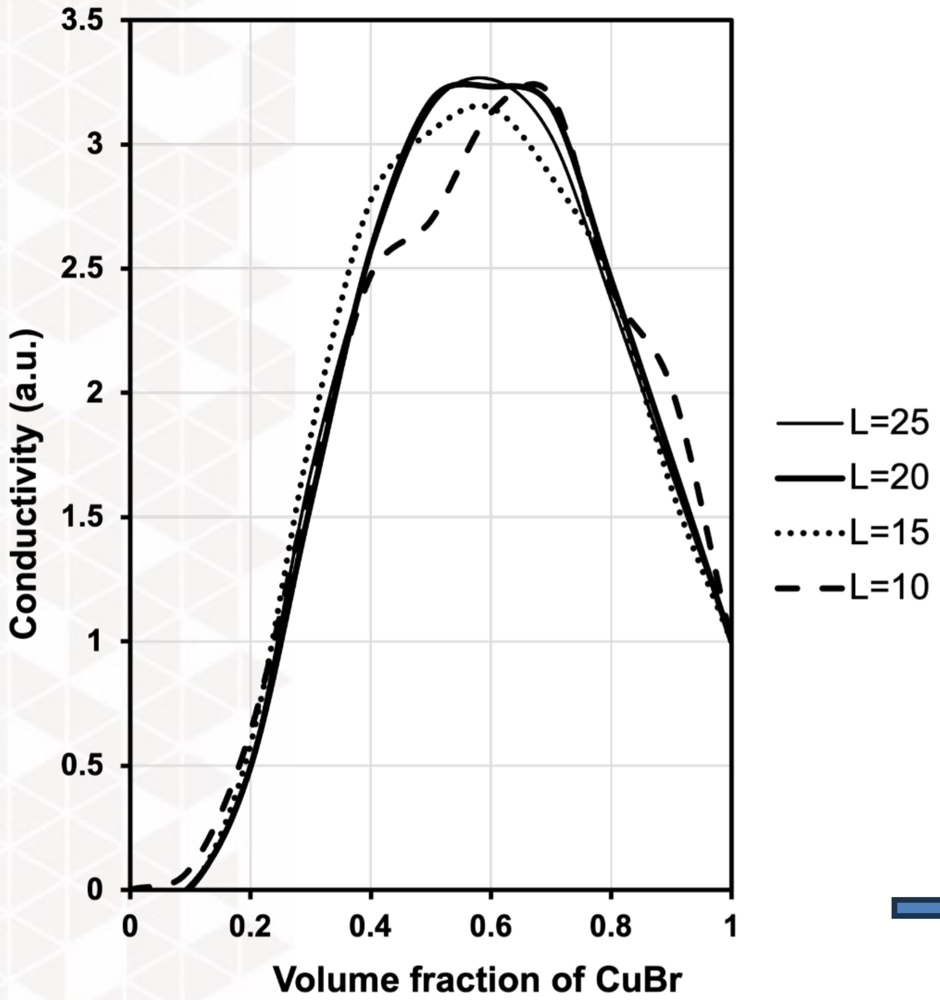
Total conduction (G_T) calculation

- Kirchoff's law applied at each node
- Total dissipated power is $P_T = G_T \cdot \Delta U^2$
- $\Delta U = 1$ V gives $P_T = G_T$

$$P_T = \sum_{i,j} g1_{i,j}\left(V_{i,j} - V_{i+1,j}\right)^2 + g2_{i,j}\left(V_{i,j} - V_{i,j+1}\right)^2 = G_T$$

III. Results and Discussion (1)

Variation of the total conductivity as a function of the volume fraction of CuBr



- L is the number of sites in one direction of the network
- g_int = 10

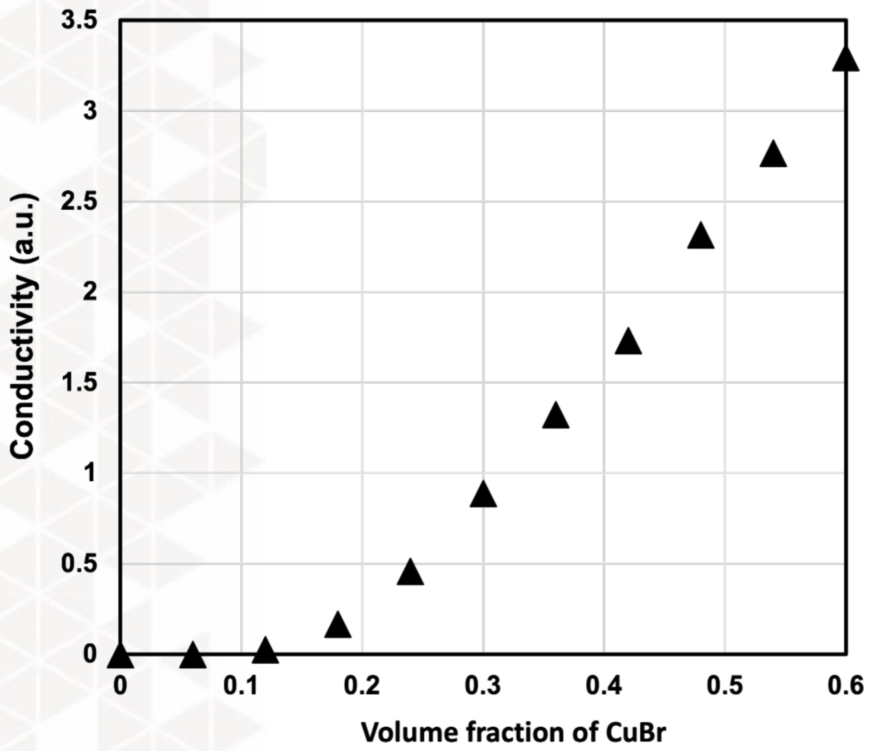
Beyond L=20, curves converge and present a bell-shape with a maximum around 0.6 volume fraction of CuBr, which agrees with previous work on composite systems [2][3].

➡ From these results, L=25 is kept in following

[2] C. Lambert and *al.*, "Temperature dependence of the resistivity in Au–$YBa_2Cu_3O_x$ sintered composites", Philosophical Magazine Part B, vol. 79, no. 7, pp. 1029–1044 (1999). [3] G. Albinet and *al.*, "Enhanced conductivity in ionic conductor-insulator composites: numerical models in two and three dimensions," The European Physical Journal B, vol. 22, pp. 421–427 (2001).

III. Results and Discussion (2)

Variation of the total conductivity as a function of the volume fraction of CuBr into a sample made of SiO_2 with a random porosity of 60%.



- 60% of porosity is introduced
- Porosity is gradually filled by CuBr

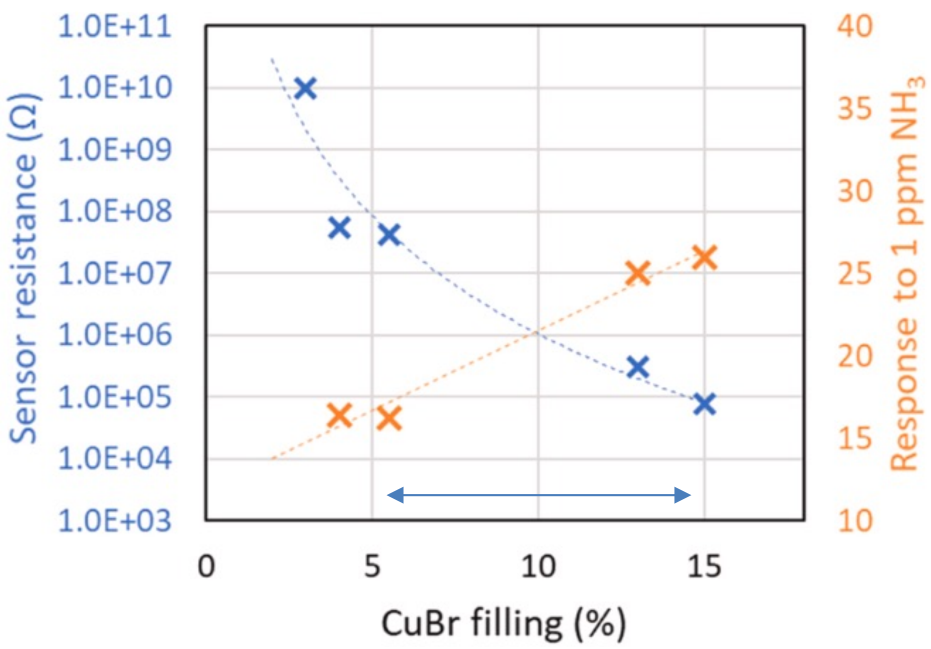
Total conductivity begins to increase around 0.18 volume fraction of CuBr

⇒ Percolation threshold, p_c ~ 0.18

III. Results and Discussion (3)

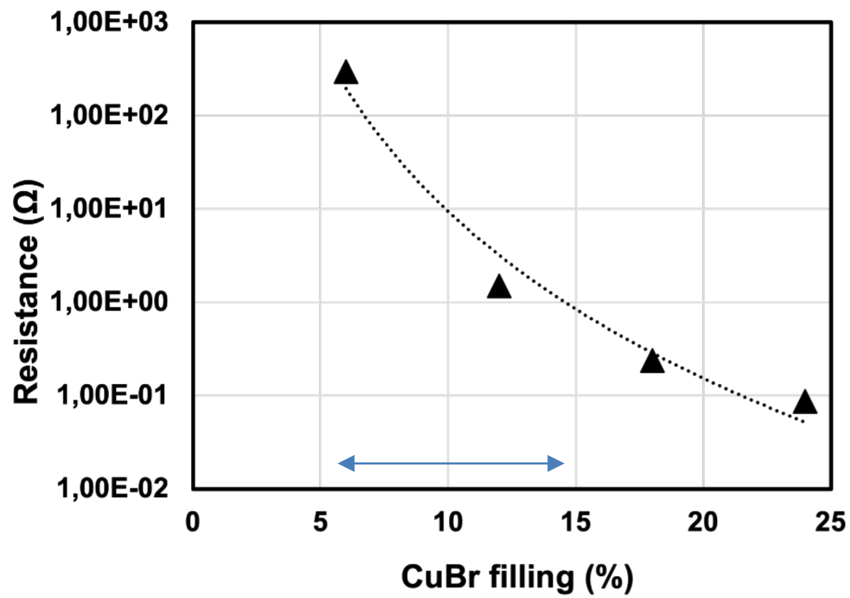
Experimental

Effect of CuBr filling on the sensor resistance and the sensor response to 1 ppm NH_3, from [1]



Calculation

Total calculated resistance in the range 0 to 25% of CuBr filling



- Same general trend
- Same order of magnitude in the range 5 -15% of CuBr filling

⇒ Good agreement between experiment and calculation

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IV. Conclusion and Future Work

Conclusion

- Worm-like structure is well modeled by random porosity

  → Build a numerical sample with worm-like porosity is not necessary

- Calculations show that less than 18% of CuBr in the total volume is enough to obtain a conducting path in the insulator SiO_2 mesoporous matrix

  → Agreement with experiment in which higher sensor performances are reached for sensitive layer composed of 13% of CuBr

Future Work

- Additional measurements, such as impedance spectroscopy coupled with calculations for other values of interface conductance (g_int) would be useful to better characterize the interfaces

Thank you for reading this far!