



30th Eurosensors Conference, EUROSENSORS 2016

Impact of the structural characteristics on the performance of light emitting capacitors using nanometric SRO multilayers fabricated by LPCVD

J. Alarcón-Salazar^{1*}, I. E. Zaldívar-Huerta¹, A. Morales-Sánchez², C. Domínguez³ and M. Aceves-Mijares¹

1 Instituto Nacional de Astrofísica Óptica y Electrónica (INAOE), Puebla 72840, México

2 Centro de Investigación en Materiales Avanzados (CIMA) S.C., Monterrey-PIIT, N.L. 66600, México

3 Institut de Microelectrònica de Barcelona, CNM-CSIC, Campus UAB, Bellaterra 08193, Spain

Abstract

This work relates the electrical, luminescent and morphological characteristics of two light emitting capacitors composed by multilayers of Silicon Rich Oxide (SRO). Multilayers alternate four conductive SRO layers (silicon excess of 12 or 14 at %) with three emitting SRO layers (silicon excess of 6 at %). Transmission electron microscopy reveals that multilayers present well-defined layers. Furthermore, it was found that layers with high silicon content induce growing of Si-nanocrystals size on layers with lower silicon excess. After the first current versus voltage measurement, electroforming produces arrays of trajectories with high silicon content. These conductive paths allow that the LEC achieves higher currents with lower voltages, preserving the emitting characteristics of SRO layers. Consequently, the electroluminescence intensity is improved as well as the blue emission depending on the conductive SRO layers.

© 2016 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Peer-review under responsibility of the organizing committee of the 30th Eurosensors Conference

Keywords: Silicon rich oxide; Silicon nanocrystals; Multilayers; Electroluminescence; Light emitting capacitors

* Corresponding author. Tel.: +52-222-266-3100; fax: +52-222-266-3100.

E-mail address: jalarcons@inaoep.mx

1. Introduction

Currently, silicon photonics is undergone an intense development in order to achieve all-silicon optoelectronic circuits. Recently, in [1] was reported the integration of a Si light source with other electro-optical devices. However, performance of the light emitter still needs to be improved. One material used to fabricate silicon light source is Silicon Rich Oxide (SRO). This material is fully compatible with silicon technology and varying the silicon excess is possible to obtain either electrical and emission properties [2, 3]. Specially, SRO deposited by Low Pressure Chemical Vapor Deposition (LPCVD) and annealed at 1100 °C during 3 hours in N₂ ambient has proved to have high emission. The most luminescent SRO has a silicon excess between 5 and 7 at %. Whether silicon excess is higher than 7 %, emission is quenched and more conductive films are achieved [2]. In the opposite, if silicon excess is less than 5 %, the material tends to be stoichiometric silicon oxide and lost its emission properties [3]. The silicon excess is controlled by the ratio, R_0 , of the reactant gases defined as $R_0 = [P_{N_2O}]/[P_{SiH_4}]$, where P_{N_2O} and P_{SiH_4} are the partial pressures of nitrous oxide and silane. The first approach to build SRO-based Light Emitting Capacitors (LECs) was to use singles layers [4]; however, electric problems affect drastically this kind of device. The main limitation is the dielectric breakdown and once it takes place the LEC is damaged, resulting not only in low reliability but also on deficient functionality. In order to avoid these problems, silicon light sources using multilayers has demonstrated to be better option, as is reported in [5, 6]. Moreover, in [7], it is shown that alternating layers with different silicon excess produces structures with superposition of properties, including better luminescence and conductivity properties. In this sense, it is mandatory to understand the influence of the morphological characteristics on the opto-electrical behavior of SRO-based light sources.

In this work, we describe the fabrication of SRO multilayers of SiO_x/SiO_y with high conduction and light emission, besides to avoid the damage to the device. Also, a statistical analysis of TEM images is addressed in order to determine sizes and densities of the silicon nanocrystals (Si-NCs) embedded in each layer. These results are related with the electrical and electroluminescent characteristics of the LEC. Depending on the conductive layer used in the ML, the main emission peak is centered in the blue or in the red region. It is shown that emission is mainly due to defects, and Si-NCs have a significant impact on the LEC conductivity.

2. Experimental procedure

For all samples, a silicon p-type (100) and 2.5-4 Ω-cm were used as substrates. They were cleaned in trichloroethylene, acetone, and deionized water in an ultrasonic bath. After that, the native oxide was removed from all samples using a buffer solution. Finally, all samples were rinsed with deionized water and spin-dried. Two types of multilayers (MLs) were deposited by LPCVD at 736 °C. The first structure alternates four conductive layers of $R_0=5$ (SRO₅) with three emitting layers of $R_0=25$ (SRO₂₅), named L-5/25. The second structure combines the same number of layers, but uses conductive layers of $R_0=10$ (SRO₁₀) instead of SRO₅, named L-10/25. Further, an extra sample of SRO₂₅ was deposited to compare between mono- and multilayer. Once all samples were deposited, they were annealed at 1100 °C during 180 min in N₂ ambient. To complete the LEC structure, a polysilicon layer with 250 nm in thickness was deposited in the top of the multilayer. Subsequently, spin on dopant (SOD) was used to N-doped the polysilicon gate, and using photolithography steps a square gate of $4.056 \pm 0.017 \times 10^{-2}$ cm² was formed.

A JOEL JEM 2200 transmission electron microscope was utilized to study the content of Si-NCs of the samples. Current versus voltage (I-V) curves were measured with a Keithley Source-Meter model 2400. Electroluminescence (EL) spectra were obtained using the same Source-Meter and a Horiba Jobin Yvon spectrometer model Fluoro-Max3.

3. Results and discussions

Fig. 1(a) shows an HRTEM image of the L-5/25. As can be seen, well-defined nanoscale layers are observed. The darkness regions correspond to SRO₅ and the surrounding regions to SRO₂₅. Likewise, inset in Fig. 1(a) exhibits the high content of Si-NCs. Two aspects have to be highlighted. First, SRO₂₅ (with 6% of silicon excess) layers have presences of Si-NCs around 1.5 nm, However Si-NCs are normally not observed in monolayers with low silicon excess, and if they are present they are small and so are their densities [8]. The second point is the high densities of

Si-NCs ($1 \times 10^{12} \text{ cm}^{-2}$) found in conductive SRO layers (SRO₅ or SRO₁₀), which influence the average size of Si-NCs in the emitting layers (SRO₂₅). Fig. 1(b) displays a comparative graph of the Si-NCs size in different samples. Clearly, a linear relationship is observed on the average size of Si-NCs on SRO₂₅ layers as function of the conductive SRO layers. Table 1 summarizes the average size and densities of Si-NCs, besides the average thickness, obtained by TEM for all samples.

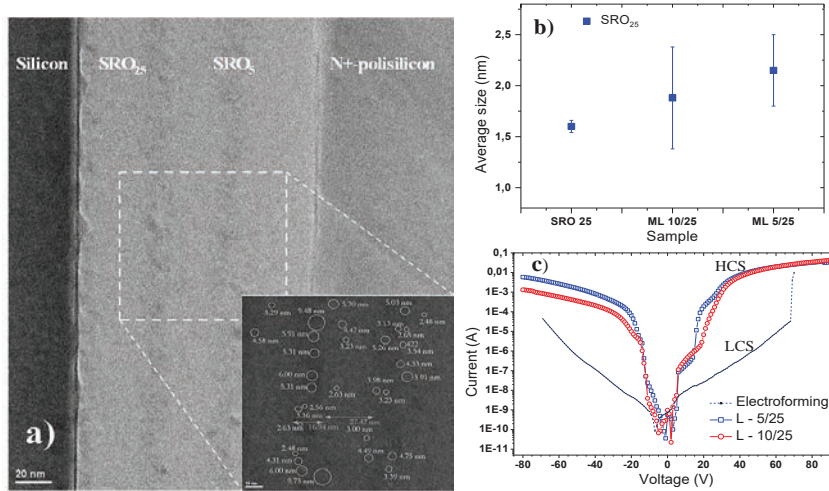


Fig. 1. (a) TEM image of a light emitting capacitor with a SRO multilayer alternating SRO₅ with SRO₂₅; (b) Average size of Si-NC embedded in SRO₂₅ layers as function of different surrounding layers; (c) Current vs voltage curves for L-5/25 and L-10/25, as can be seen very similar curves are obtained for either devices.

Table 1. Average thickness, size and densities of Si-NCs in each SRO layer.

Sample	Average size of Si-NCs [nm]			Average thickness [nm]			Densities of Si-NCs [cm ⁻²]		
	SRO ₅	SRO ₁₀	SRO ₂₅	SRO ₅	SRO ₁₀	SRO ₂₅	SRO ₅	SRO ₁₀	SRO ₂₅
L-5/25	4.01 ± 1.53	---	2.15 ± 0.35	14.5 ± 5.1	---	25.7 ± 1.7	1.06x10 ¹²	---	5.79x10 ¹¹
L-10/25	---	3.61 ± 1.48	1.86 ± 0.51	---	11.8 ± 2.1	26.0 ± 0.9	---	1.30x10 ¹²	7.43x10 ¹¹
SRO ₂₅	---	---	1.60 ± 0.06	---	---	33.1 ± 0.4	---	---	~10 ¹⁰

Fig. 1(c) shows the I-V curve for both devices. In fresh samples, the devices have a Low Conduction State (LCS), which is lost once the devices are tested, achieving a High Conduction State (HCS). This behavior is reproduced in the subsequent measurements. This effect was reported in [9], where trajectories with high silicon content are formed due to an electrical treatment (electroforming). Therefore, we assume that electroforming phenomena take place in the multilayers due to a wide range of Si-NCs sizes and their high densities (see Table 1). The size and the density propitiate that electrical conductivity increases through the multilayer. In [7], it is explained these phenomena by the space charge limited and trap-assisted tunneling mechanisms, thereby carriers can overcome a barrier height or tunneling from one trap to another.

Fig. 2 exhibits the normalized EL spectra for both LECs. Devices were forward biased (inversion mode). As can be observed, for low electric field one peak centered in the red region is observed; however as the electric field is increased two emission peaks are defined, finding that L-5/25 has higher emission in the blue region and L-10/25 in the red region. Neutral oxygen defects (NOVs) and non-bringing oxygen hole centers (NBOHCs) are associated to the emission in the blue and red regions, respectively [7]. Both defects have been found into emitting SRO layers [3]. Then, in the L-5/25 more conductive layers (SRO₅) produce that electrons obtain enough energy to reach high energy blue emissive center in the SRO₂₅ layers. For ML 10/25, in order to stimulate blue centers higher fields are required; however, too high fields applied to the ML could damage the device.

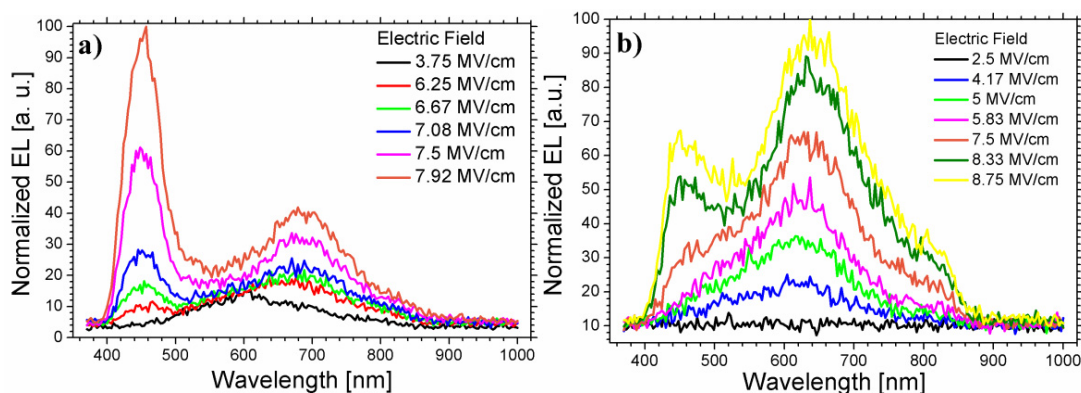


Fig. 2. Normalized EL spectra for (a) L-5/25 and (b) L-10/25. Different emission peaks are observed depending on the SRO conductive layer.

4. Conclusions

Two light emitting capacitors based on multilayer of SRO (5/25 and 10/25) were fabricated and studied. It was found that SRO with 6 % of silicon excess (SRO₂₅) presents Si-NCs from 1.6 to 2.1 nm, and this value depends on the surrounding material. In case of SRO₅ and SRO₁₀, average size of 4.01 nm and 3.61 nm were determined. Likewise, average densities of Si-NCs in the order of 10¹¹ and 10¹² cm⁻² were found on emitting and conductive SRO layers, respectively. Both LECs presents an electrical behavior labeled as HCS. EL spectra shows two emission peaks, but blue emission dominates in L-5/25 and red emission in L-10/25. These results are related to the conductive SRO layers; thereby SRO₅ allows exciting more blue centers in the SRO₂₅ than SRO₁₀.

Acknowledgements

The authors appreciate the economic support of CONACyT, México, and J. Alarcón-Salazar is grateful for his scholarship No. 353251. Likewise, authors recognize to Pablo Alarcón, Victor Aca and Armando Hernández for the help during the fabrication process.

References

- [1] A. A. González-Fernández, J. Juvert, M. Aceves-Mijares and C. Domínguez, Monolithic Integration of a Silicon-Based Photonic Transceiver in a CMOS Process, *IEEE Photonics J.* 8(1); 2016, 7900213.
- [2] J. A. Luna-López, M. Aceves-Mijares, J. Carrillo-López, and A. Morales-Sánchez, Photoconduction in silicon rich oxide films obtained by low pressure chemical vapor Deposition, *J. Vac. Sci. Technol. A* 28; 2010.
- [3] M. Aceves-Mijares, A. A. González-Fernández, R. López-Estopier, A. Luna-López, D. Berman-Mendoza, A. Morales, C. Falcony, C. Domínguez, and R. Murphy-Arteaga On the Origin of Light Emission in Silicon Rich Oxide Obtained by Low-Pressure Chemical Vapor Deposition 890701.
- [4] J. Alarcón-Salazar, M. A. Vázquez-A., E. Quiroga-González, I. E. Zaldívar-Huerta and M. Aceves-Mijares, Si/SRO/Poly-Si MOS-like devices with enhanced light emission through ionic Si surface roughening, in press.
- [5] E. Quiroga-González, W. Bensch, M. Aceves-Mijares, Z. Yu, R. López-Estopier, K. Monfil-Leyva, On the photoluminescence of multilayer arrays of silicon rich oxide with high silicon content prepared by low pressure chemical vapor deposition, *Thin Solid Films* 519; 2011, 8030–8036.
- [6] Sheng-Wen Fu, Hui-Ju Chen, Hsuan-Ta Wu, Chuan-Feng Shih, Effect of SiO₂ layers on electroluminescence from Si nanocrystal/SiO₂ superlattices prepared using argon ion beam assisted sputtering, *Vacuum* 126; 2016, 59-62.
- [7] J. Alarcón-Salazar, I. E. Zaldívar-Huerta and M. Aceves-Mijares, Electrical and electroluminescence characterization of nanometric multilayers of SiOx/SiOy obtained by LPCVD including non-normal emission. *J. Appl. Phys.* 119; 2016, 215101.
- [8] J. A. Luna-López, A. Morales-Sánchez, M. Aceves-Mijares and C. Domínguez, Analysis of surface roughness and its relationship with photoluminescence properties of silicon-rich oxide films, *J. Vac. Sci. Technol. A* 27; 2009.
- [9] Jun Yao, Lin Zhong, Douglas Natelson and James M. Tour. In situ imaging of the conducting filament in a silicon oxide resistive switch. *Scientific Reports* 2; 2012; 242.