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A new Model for Prediction of luminescent compartment of nanostructured Silicon Rich Oxide

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Abstract*—A new theoretical model is presented to describe a set of chemical reactions that can potentially occur during the process of obtaining silicon rich oxide (SRO) films, an outside stoichiometry material, regardless of the technique used to grow such films. Particularly, chemical reactions that occur during the process of growing of SRO films by LPCVD technique are emphasized in the model presented. We include the effects of annealing reactions which are present when SRO films are annealed. We suggest and evaluate either some types of molecules or resulting nanostructures and we predict theoretically, by applying the density functional theory, the contribution that they may have to the phenomenon of luminescence which is measured in SRO films. Also, we have calculated the opto-electronic properties of SRO films. The suggested model provides enough information required to propose different silicon formed nano-agglomerates.

***Index Terms*—Global Reaction Model, luminescence, Silicon rich-Oxide, LPCVD.**

I.-Background

The motivation of this work is to introduce a new model, which we had called the Global Reaction Model (GRM), for the theoretical study of the optical and electronics properties of Silicon Rich Oxides (SRO) structures regardless of the technique used to fabricate such structures. For this, we have reviewed firstly some important experimental results about measurements of structural and optical properties carried out on SRO samples grown by different techniques and the theoretical models available in order to describe a SRO network. The aim of this review is threefold; on the one hand, we found relevant information in relationship to actual quantification of silicon nano-clusters (Si-nCs) about their size, electromagnetic range of emission, molecular structure and important parameters which are responsible for making variations of optical properties of SRO.

On the other hand, we take this experimental information as background in order to focus correctly on our theoretical research predicted by using the Density Functional Theory (DFT) method corresponding to atomic composition of different silicon isomers suggested simulating the Si-nCs embedded in SRO films. Finally, we have developed a set of equations for describing completely the phenomenon under study as foundation of the proposed GRM.

Silicon Rich Oxide (SRO) thin films have been studied extensively and are very interesting due to their opto-electronic properties, particularly those related with luminescence. In general, luminescent properties can provide significant information regarding the crystalline structure of a material and, in the case of SRO thin films, electro-luminescence properties are particularly important since these films can be used to fabricate luminescent devices[1].

Today, a few models are frequently used to describe a SRO network, namely: the Mixture Model (MM)[2], the Random Bonding Model (RBM)[3] and the Intermediate Model (IM)[4] introduced in 2011 by Novikov and Gritsenko. In 2012 Davor et al.[5], in an extensive review, considered that the actual structure seems to be greatly determined by the deposition procedure. In some works, the SiOx structures films obtained by radio-frequency Sputtering and physical evaporation were claimed to correspond to RBM, whereas the SiOx films obtained by magneto-sputtering, Plasma Enhanced Chemical Vapor Deposition (PECVD), have been assigned to MM. IM was used to describe SiOx layers prepared by LPCVD using SiH_4 and N_2O as a reactant precursors at 750 °C. The

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Volume 4, Issue 2, March 2015

models currently found in the literature are oriented to evaluate the changes in nanostructured thin films derived from specific deposition procedures.

This paper is outlined as follows: in Methods section we present the basic equations of chemical reactions of our model considering four subsections, namely, Global and Partial Reactions as well as Annealing Reactions and Side-way or Secondary reactions and Formation of siloxanes derivates. In section Results and Discussion we present different PL spectra of agglomerates of the several molecular structures studied in this work. Finally, in Section of Conclusions we stress the main contributions of this paper.

II. METHODS

A. Global and Partial reaction(s)

We present for first time a new model, which consider the Global and Partial Reaction(s) necessary to generate the oxide matrices (SiO_2, Si_2O_3, SiO and Si_2O), the annealing reactions for explaining the compositional changes after and before the thermal treatment and consequently the changes in luminescence spectra intensity and a set of secondary reactions of the oxide matrixes with the hydrogen produced to obtain the ions that could be associated to the emission in SRO thin films with specific defects.

When SRO is prepared by Low Pressure Chemical Vapor Deposition (LPCVD) technique, a gas mixture of N_2O and SiH_4 is habitually used [6,7] and the Si excess content can be modified by the gas flow ratio Ro = $[N_2O]/[SiH_4]$[8,9]. The silicon excess can be as high as 17 at. % for Ro = 3; and experimentally stoichiometric SiO_2 (a non-free silicon film) can be obtained for Ro ≥ 50[10]. Theoretically, Ro = 40 corresponds to the stoichiometric silica, when a mixture silane-nitrogen at 5 at. % is used. Experimentally, there is enough evidence that SRO thin films are constituted by a silicon oxides mixture rather than by only one of them, regardless of the value of Ro, therefore the global reaction is constituted by

$$2N_2O_{(gas)} + 19N_{2(gas)} + SiH_{4(gas)} \rightleftarrows (1-x)SiO_{2(solid)} + xSi_{solid} + xO_{2(gas)} + 2H_{2(gas)} + 21N_{2(gas)}, \quad (1)$$

Where de ratio Ro is given by

$$Ro = \frac{2N_2O_{(gas)}}{SiH_{4(gas)} * \frac{SiH_{4(gas)}}{19N_{2(gas)}+SiH_{4(gas)}}} = \frac{2}{1 * \frac{1}{19+1}} = 40,$$

Being *x* the quantity in moles of Si_{solid} produced by the reaction (1).In the case of Ro=0, it means a reaction in absence of oxygen according with reaction (2),

$$wSiH_{4(gas)} \rightleftharpoons wSi_{solid} + 2wH_{2(gas)}, \quad (2)$$

Where *w* is the quantity in moles of Si_{solid} produced by the reaction (2).When the ratio Ro assumes the value Ro=20, the reaction could be as shown by (3)

$$N_2O_{(gas)} + 19N_{2(gas)} + SiH_{4(gas)} \rightleftarrows (1-y)SiO_{(solid)} + ySi_{solid} + yO_{2(gas)} + 2H_{2(gas)} + 20N_{2(gas)}, \quad (3)$$

And the corresponding ratio Ro given by

$$Ro = \frac{N_2O_{(gas)}}{SiH_{4(gas)} * \frac{SiH_{4(gas)}}{19N_{2(gas)}+SiH_{4(gas)}}} = \frac{1}{1 * \frac{1}{19+1}} = 20.$$

We observe that in Eq. (3) the parameter *y* is the quantity in moles of Si_{solid} produced in such reaction. For Ro=10, we postulate the reaction (4) as follows:

ISSN: 2319-5967
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$$N_2O_{(gas)} + 38N_{2(gas)} + 2SiH_{4(gas)} \rightleftarrows (1-z)Si2O_{(solid)} + 2zSi_{solid} + (0.5*z)O_{2(gas)} + 4H_{2(gas)} + 39N_{2(gas)}, \quad (4)$$

and

$$Ro = \frac{N_2O_{(gas)}}{2SiH_{4(gas)} * \frac{2SiH_{4(gas)}}{38N_{2(gas)}+2SiH_{4(gas)}}} = \frac{1}{2*\frac{2}{38+2}} = 10.$$

Where z in Eq. (4) is the quantity in moles of the middle of Si_{solid} produced by the reaction (4). Finally, for Ro=30, the chemical reaction is given by Eq. (5):

$$3N_2O_{(gas)} + 38N_{2(gas)} + 2SiH_{4(gas)} \rightleftarrows (1-v)Si2O_{3(solid)} + 2vSi_{solid} + (1.5*v)O_{2(gas)} + 4H_{2(gas)} + 41N_{2(gas)}, \quad (5)$$

And

$$Ro = \frac{3N_2O_{(gas)}}{2SiH_{4(gas)} * \frac{2SiH_{4(gas)}}{38N_{2(gas)}+2SiH_{4(gas)}}} = \frac{3}{2*\frac{2}{38+2}} = 30.$$

In Eq. (5), v is the quantity in moles of the middle of Si_{solid} produced by the reaction (5). Considering the partial reactions from Eq.(1) to Eq.(5), the global reaction is determined by Eq.(6):

$$\begin{aligned} &2N_2O_{(gas)} + 19N_{2(gas)} + SiH_{4(gas)} + wSiH_{4(gas)} + N_2O_{(gas)} + 19N_{2(gas)} + SiH_{4(gas)} \\ &+ N_2O_{(gas)} + 38N_{2(gas)} + 2SiH_{4(gas)} + 3N_2O_{(gas)} + 38N_{2(gas)} \\ &+ 2SiH_{4(gas)} \\ &\rightleftarrows (1-x)SiO_{2(solid)} + xSi_{solid} + xO_{2(gas)} + 2H_{2(gas)} + 21N_{2(gas)} \\ &+ wSi_{solid} + 2wH_{2(gas)} + (1-y)SiO_{(solid)} + ySi_{solid} + yO_{2(gas)} \\ &+ 2H_{2(gas)} + 20N_{2(gas)} + (1-z)Si_2O_{(solid)} + 2zSi_{solid} + 0.5*zO_{2(gas)} \\ &+ 4H_{2(gas)} + 39N_{2(gas)} + (1-v)Si_2O_{3(solid)} + 2vSi_{solid} + 1.5*vO_{2(gas)} \\ &+ 4H_{2(gas)} + 41N_{2(gas)}. \end{aligned} \quad (6)$$

Simplifications in Eq.(6) lead to

$$\begin{aligned} &7N_2O_{(gas)} + 114N_{2(gas)} + (6+w)SiH_{4(gas)} \\ &\rightleftarrows (1-x)SiO_{2(solid)} + (x+y+2z+2v+w)Si_{solid} \\ &+ (x+y+0.5*z+1.5*v)O_{2(gas)} + (12+2w)H_{2(gas)} + (121)N_{2(gas)} \\ &+ (1-y)SiO_{(solid)} + (1-z)Si_2O_{(solid)} + (1-v)Si_2O_{3(solid)}. \end{aligned} \quad (6.1)$$

On the other hand, we define a global ratio RoG as follows:

$$RoG = \frac{7N_2O_{(gas)}}{(6+w)SiH_{4(gas)} * \frac{(6+w)SiH_{4(gas)}}{114N_{2(gas)}+(6+w)SiH_{4(gas)}}},$$

$$RoG = \frac{7*(114+6w)}{(6+w)^2}. \quad (6.2)$$

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When SRO films are obtained by LPCVD technique, *w=0*, and the global reaction described by equation (6) is simplified to Eq. (7):

$$7N_2O_{(gas)} + 114N_{2(gas)} + 6SiH_{4(gas)} \rightleftarrows (1-x)SiO_{2(solid)} + (x+y+2z+2v)Si_{solid} + (x+y+0.5*z+1.5*v)O_{2(gas)} + 12H_{2(gas)} + 121N_{2(gas)} + (1-y)SiO_{(solid)} + (1-z)Si_2O_{(solid)} + (1-v)Si2O_{3(solid)}. \quad (7)$$

$(SRO)_{solid}$ thin films in equilibrium are always silicon oxides mixtures constituted by:

$$(1-x)\ SiO_{2(solid)}, (x+y+2z+2v)Si_{(solid)}, (1-y)\ SiO_{(solid)}, (1-z)Si_2O_{(solid)}\ and\ (1-v)\ Si_2O_{3(solid)}.$$

And, $RoG = \frac{7*114}{36} = 22.167$.

Boundary conditions are imposed as follows:

$$0 \leq Si_{solid} \leq x+y+2z+2v \leq 1.0$$
$$0 \leq x \leq 1.0$$
$$0 \leq y \leq 1.0$$
$$0 \leq z \leq 1.0$$
$$0 \leq v \leq 1.0$$

The theoretical composition of SRO thin films will be:

$$SiO_{2(solid)} = \frac{1-x}{4+z+v} \quad (8.1)$$

$$Si_2O_{3(solid)} = \frac{1-v}{4+z+v} \quad (8.2)$$

$$SiO_{(solid)} = \frac{1-y}{4+z+v} \quad (8.3)$$

$$Si_2O_{(solid)} = \frac{1-z}{4+z+v} \quad (8.4)$$

$$Si_{solid} = \frac{x+y+2z+2v}{4+z+v} \quad (8.5)$$

B. Annealing reactions

When SRO films are deposited using the LPCVD technique, experimentally has been probed that resultant nanostructures cannot emit in the visible, or the emission intensity in the visible region is limited, depending on the parameter Ro. As a consequence of the heat treatment of these films, it has been experimentally shown also, that emission appears in the visible region of electromagnetic spectra or the observed emission in visible region intensifies. In the proposed model, we suggest strongly that when SRO thin films are annealed, some oxides are degraded. The plausible "annealing reactions" proposed are:

$$SiO_{2(solid)} + Heat \rightleftarrows (1-\xi)SiO_{(solid)} + (0.5+0.5\xi)O_{2(gas)} + \xi Si_{solid}, \quad (9)$$

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Volume 4, Issue 2, March 2015

$$Si_2O_{3(solid)} + Heat \rightleftarrows 2(1-\xi')SiO_{(solid)} + (0.5+\xi')O_{2(gas)} + 2\xi' Si_{solid}, \quad (10)$$

$$Si_2O_{3(solid)} + Heat \rightleftarrows (1-\xi'')Si_2O_{(solid)} + (1+0.5\xi'')O_{2(gas)} + \xi'' Si_{solid}, \quad (11)$$

Double arrow stands for denoting equilibrium condition, ξ , ξ' and ξ'' are the progress parameters of the annealing reactions. The extent of progress of reaction is defined as the ratio between the total change in the number of moles of a species and their stoichiometric coefficients.The numerical value of these parameters will depend on time, temperature and flow of inert gas employed during the thermal process.

C. Side-way or Secondary Reaction

As it is known, a mole of any substance contains the number of atoms equal to the Avogadro's number. In order to perform first-principles calculations, the problem must be modeled with a maximum of 100 atoms. Equations from Eq.(8.1) to Eq.(8.4) must be rewritten in terms of the number of atoms, leaving the following oxides:

$$\mathrm{Si}_n O_{2n}\ , Si_{2n}O_{3n}, \mathrm{Si}_n O_n\ , Si_{2n}O_n. \quad (12)$$

The reactor effluent is a gaseous mixture of hydrogen, oxygen and nitrogen. From Eq. (7) we see that the effluent gas mixture is formed by:

$$(x + y + 0.5 * z + 1.5 * v)O_{2(gas)} + 12H_{2(gas)} + 121N_{2(gas)}.$$

The hydrogen gaseous can react according to,

$$H_2 = [H]^+ + [H:]^-.$$

The hydrogen ion formed, in turn, reacts with the silicon oxides to form ions as follows:

$$[\mathrm{Si}_n O_{2n}] + [H]^+ \rightleftarrows [\mathrm{Si}_n O_{2n} H]^+ (13)$$

$$[\mathrm{Si}_n O_{2n} H]^+ + [H]^+ \rightleftarrows [\mathrm{Si}_n O_{2n} H_2]^{2+} (14)$$

$$[\mathrm{Si}_{2n} O_{3n}] + [H]^+ \rightleftarrows [\mathrm{Si}_{2n} O_{3n} H]^+ (15)$$

$$[\mathrm{Si}_{2n} O_{3n} H]^+ + [H]^+ \rightleftarrows [\mathrm{Si}_{2n} O_{3n} H_2]^{2+} (16)$$

$$[\mathrm{Si}_n O_n] + [H]^+ \rightleftarrows [\mathrm{Si}_n O_n H]^+ (17)$$

$$[\mathrm{Si}_n O_n H]^+ + [H]^+ \rightleftarrows [\mathrm{Si}_n O_n H_2]^{2+} (18)$$

$$[\mathrm{Si}_{2n} O_n] + [H]^+ \rightleftarrows [\mathrm{Si}_{2n} O_n H]^+ (19)$$

$$[\mathrm{Si}_{2n} O_n H]^+ + [H]^+ \rightleftarrows [\mathrm{Si}_{2n} O_n H_2]^{2+} (20)$$

During heat treatment, the reactions which correspond to Eqs.(14), (16), (18) and (20) produce a dehydration of these cations, resulting in the formation of new ones, as follows:

$$[\mathrm{Si}_n O_{2n} H_2]^{2+} \rightleftarrows [\mathrm{Si}_n O_{2n-1}]^{2+} + H_2O \uparrow \quad (21)$$

$$[\mathrm{Si}_{2n} O_{3n} H_2]^{2+} \rightleftarrows [\mathrm{Si}_{2n} O_{3n-1}]^{2+} + H_2O \uparrow \quad (22)$$

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ISO 9001:2008 Certified
International Journal of Engineering Science and Innovative Technology (IJESIT)
Volume 4, Issue 2, March 2015

$$[Si_nO_nH_2]^{2+} \rightleftarrows [Si_nO_{n-1}]^{2+} + H_2O \uparrow \quad (23)$$

$$[Si_{2n}O_nH_2]^{2+} \rightleftarrows [Si_{2n}O_{n-1}]^{2+} + H_2O \uparrow \quad (24)$$

The moieties obtained from reactions described by equations from (20) to (23) are:

$$[Si_nO_{2n-1}]^{2+};\ [Si_{2n}O_{3n-1}]^{2+};\ [Si_nO_{n-1}]^{2+};[Si_{2n}O_{n-1}]^{2+}. \quad (25)$$

Oxides described by Eq.(12) may react with hydrogen in a different way as that previously described in reactions from Eq.(13) to Eq.(20), forming anions according to the following reactions:

$$[Si_nO_{2n(solid)}] + 2H_{2(gas)} \rightleftarrows [Si_{n-1}O_{2n(solid)}]^{4-} + SiH_{4(gas)}, \quad (26)$$

$$[Si_{2n}O_{3n(solid)}] + 2H_{2(gas)} \rightleftarrows [Si_{2n-1}O_{3n(solid)}]^{3-} + SiH_{4(gas)}, \quad (27)$$

$$[Si_nO_n] + 2H_{2(gas)} \rightleftarrows [Si_{n-1}O_{n(solid)}]^{2-} + SiH_{4(gas)}, \quad (28)$$

$$[Si_{2n}O_n] + 2H_{2(gas)} \rightleftarrows [Si_{2n-1}O_{n(solid)}]^{1-} + SiH_{4(gas)}. \quad (29)$$

The anions formed from the reactions (26) to (29), are different oxides matrices containing vacancies or defects of silicon which may or not be present in the films of SRO.

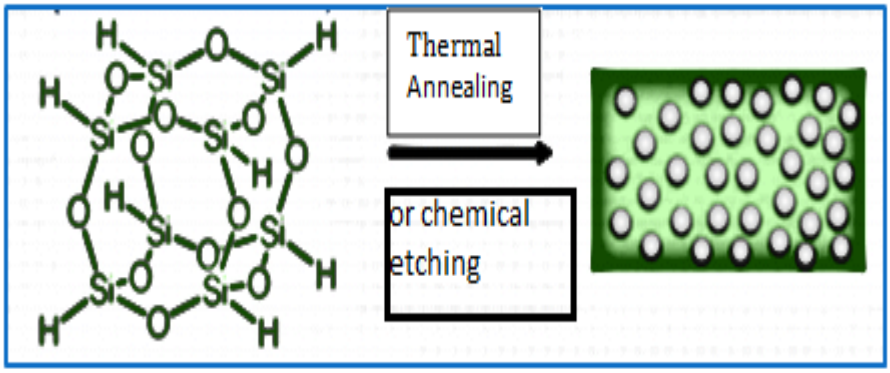
D. Formation of siloxanes derivates.

Henderson et al. [11], have reported that the preparation of SiO_2-embedded silicon nanocrystals (Si-NCs) from the thermal processing of sol−gel polymers could be derived from trichlorosilane ($HSiCl_3$). Straightforward addition of water to $HSiCl_3$ generates a cross-linked $(HSiO_{1.5})_n$ sol−gel polymer suitable for the generation of bulk quantities of SiO_2-embedded Si-NCs.The structural differences between the network structure $2(HSiO_{1.5})_n$ polymer and hydrogen silsesquioxane (HSQ, $(HSiO_{1.5})_{2n}$) result in controllable differences in the resulting oxide-embedded Si-NCs produced from these precursors. A well-known fact is that SRO films can be obtained with LPCVD technique employing silane and nitrous oxide. In the GRM we consider that the reaction of these gases could produce HSQ through Eq. (29) in equilibrium with branched network structure (BNS) as displayed in Eq. (30).

$$n[SiH_{4(gas)}] + 1.5\ n(N_2O_{(gas)}) \rightarrow 0.5(HSiO_{1.5})_{2n\ (solid)} + 1.5\ n(N_2 + H_2)_{(gas)} \quad (30)$$

$$(HSiO_{1.5})_{2n\ (solid)} \rightleftarrows 2(HSiO_{1.5})_{n\ (solid)} \quad (31)$$

Figure 1 displays the formation of siloxane derivates. Freestanding Si-NCs could be obtained through chemical etching and/or thermal treatment of the oxide matrix and exhibit tunable PL throughout the visible spectrum. HSQ should produce bigger silicon nano-agglomerates than BNS. Likewise, the present Si-NCs should exhibit size-dependent photoluminescence (PL) in accordance with the principles of quantum confinement.



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International Journal of Engineering Science and Innovative Technology (IJESIT)
Volume 4, Issue 2, March 2015

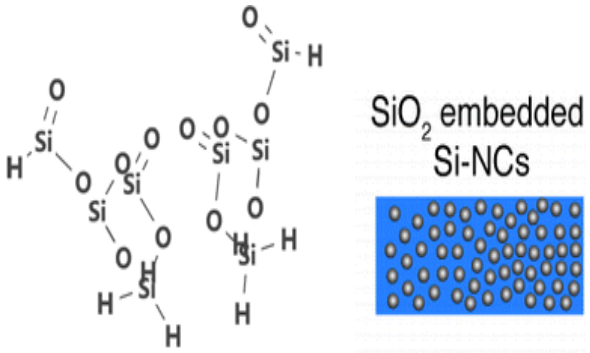

Fig 1Siloxane derivatives for n=4. Cage structure of hydrogen silsesquioxane $(HSiO_{1.5})_{2n}$ (top), and with two branched network structures $2(HSiO_{1.5})n$ (bottom).

III.RESULTS AND DISCUSSION

A. Size Effect on Silicon isomers agglomerates as responsible for luminescence suggested simulating the Si-nCs embedded in SRO films.

Various techniques have been reported for synthesizing silicon quantum dots (Si QDs) embedded in an oxide matrix. The widely used techniques include (PECVD) [12,13], ion implantation [14], and magnetron sputtering [15] among others. The most common method to form Si QDs entails deposition of a thick Si-rich oxide (SRO (SiO_x, $x < 2$)) monolayer.

In Figure 2 we have displayed the calculated UV Vis spectra for silicon agglomerates with different sizes, from Si_7 to Si_{20}. In this work we consider all isomers from each agglomerate size found in literature, and exhibit only the spectra corresponding to isomers with the lowest energy, i.e., the most stable structure. With some exceptions, we can say that silicon agglomerates will exhibit luminescence in the range of visible region. Exceptionally, only the silicon agglomerate (Si_{7A}) shows emission in ultra violet region, more exactly at 382.41 nm. Apart from this, we also evaluated four isomers, namely, isomers from Si_{7A} to Si_{7D}. We found two values for calculated maximum emissions, which are not shown, they were: isomer A (at 382.41 nm), isomer B (at 660.93 nm), isomer C (598.11 nm) and isomer D (591.98 nm) which corresponds with the most stable configuration. It is worth noting that isomer 15A displays only one wide band emission having a maximum value at 525.31 nm. On the other hand, isomer Si_{8E} results with a second incipient band-like shoulder (at 535.14 it is found the highest peak of emission and around at 642.60 nm is located the incipient emission, respectively). As far as the size of agglomerate increases, there is a redshift color.

Two wide bands overlapped in the range from 450 to 600 nm have resulted for the case of low energy isomers which correspond to agglomerates 9C, 10A, 14A, 16B and 20A.For isomer 11E the two wide bands are clearly separated, the first one remains in visible region (524.32 nm), but the second one is displaced to NIR (834.86 nm). Respect to isomers 12E, 18A and 19B, they present multiple bands covering all visible region, whereas isomer 17C, with manifold bands also, exhibits luminescence in the red region and NIR.

We stress that isomer 13A has a complex behavior, as shown in Figure 2, we can appreciate two main wide bands well defined at 744.14 nm and 1040.06 nm and adjacently to each one we identify two smaller bands located around at 670.65 nm and 1098.66 nm, the four bands lie in UV VIS calculated spectrum, two of them overlapped in visible region and the others, overlapped also, in NIR. The highest intensity values were observed at 744.14 nm and 1040.06 nm. We have highlighted in red color for silicon agglomerates 17C (middlemost wide band) and 20A (right wide band) numerical intervals instead of punctual values due to we found doublets in these emissions wavelength intervals.

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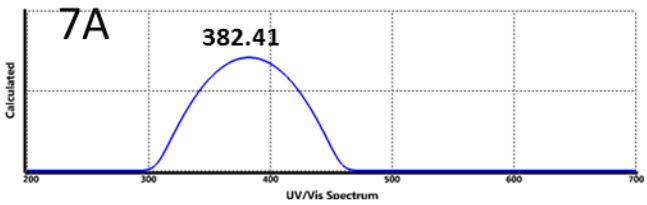

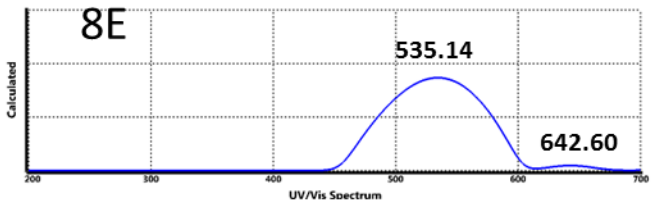

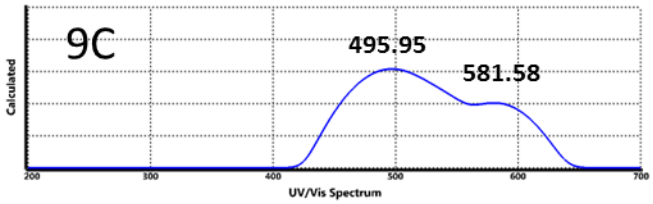

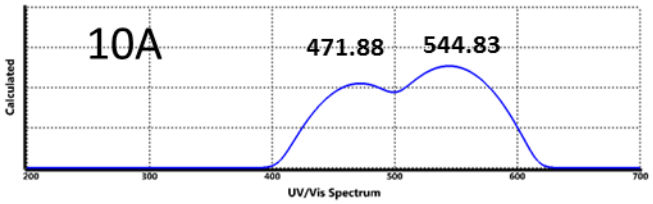

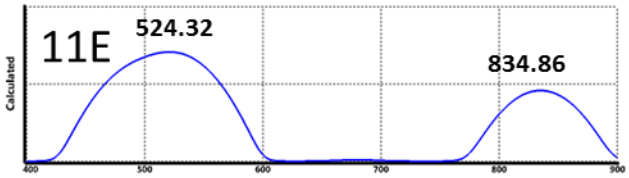

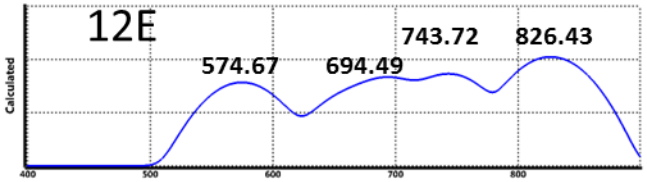

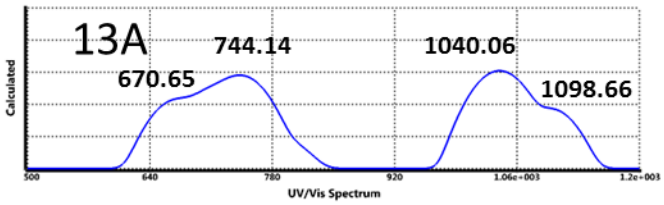

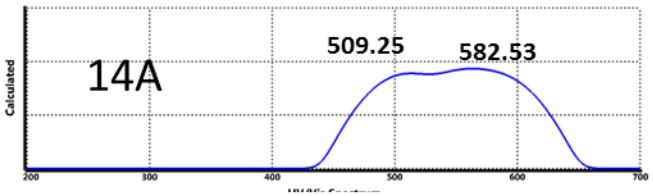

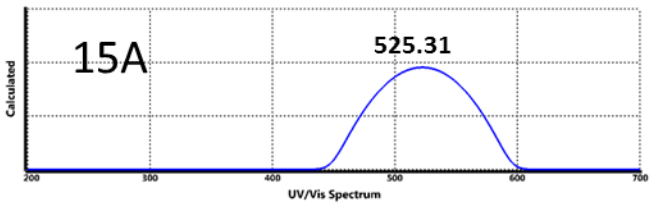

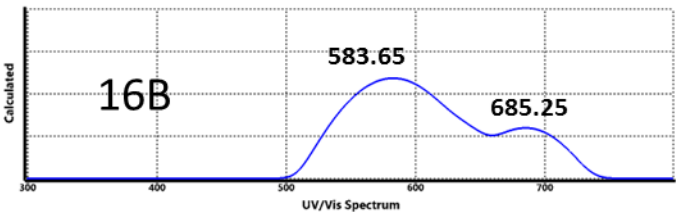

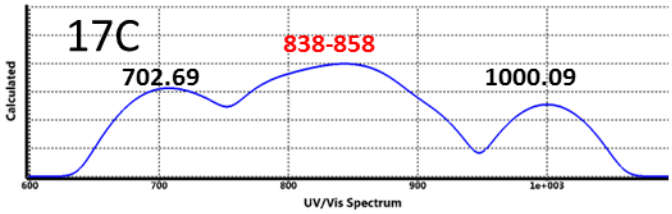

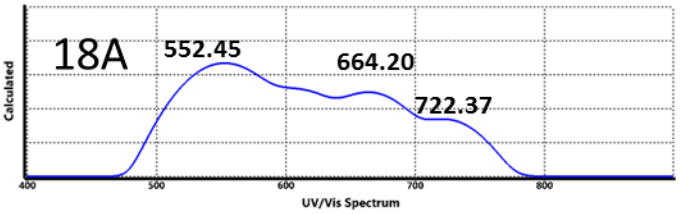

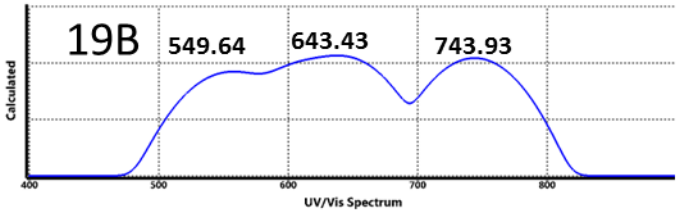

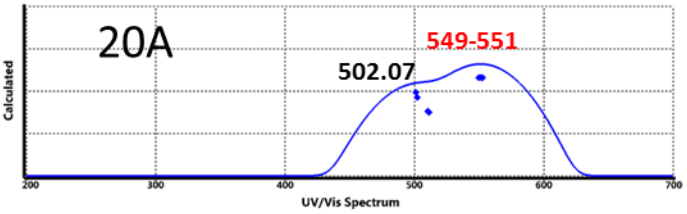

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IV. CONCLUSION

We have carried out the implementation of a new model called the Global Reaction Model in order to predict the theoretical luminescent behavior of nanostructured Silicon Rich Oxide films which are considered as constituted by nanostructures. With this model, we have also predicted de existence of silsesquioxanes (nanostructures constituted by two cyclosiloxanes joined by oxygen bridges connecting pairs of silicon atoms) which may be present in the formation of Si-NCs. Besides, the proposed model is successfully applied to evaluate, the contribution to the luminescent phenomenon of silicon nano-agglomerates, considering the impact of the size of the nanostructure. The results are shown for agglomerates from 7 to 20 silicon atoms considering the isomers more stable. Such isomers exhibit luminescence in the visible region. It is worth noting that only the isomer Si_{7A} have shown luminescence in the ultraviolet region at 382.41 nm. It is found that isomer Si_{11E} exhibited two well defined wide bands one peaked at 524.32 nm and the other at 834.86 nm. All others isomers showed overlapped bands in different regions of the visible spectrum.

V. CONFLICT OF INTERESTS

The authors declare that there is no conflict of interests regarding the publication of this paper.

VI. ACKNOWLEDGMENT

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