

Structural and Optical Properties of Cu^{2+} + Ce^{3+} Co-Doped ZnO by Solution Combustion Method

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How to cite this paper: López-Romero, S., Jiménez, M.J.Q. and García-Hipólito, M. (2016) Structural and Optical Properties of Cu^{2+} + Ce^{3+} Co-Doped ZnO by Solution Combustion Method. *World Journal of Condensed Matter Physics*, 6, 300-309.

<http://dx.doi.org/10.4236/wjcmp.2016.64029>

Received: September 6, 2016

Accepted: November 27, 2016

Published: November 30, 2016

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Abstract

In this work, ZnO, Ce^{3+} doped ZnO ($\text{ZnO}/\text{Ce}^{3+}$) and Cu^{2+} + Ce^{3+} co-doped ZnO ($\text{ZnO}/\text{Cu}^{2+}$ + Ce^{3+}) solid solutions powders were synthesized by a solution combustion method maintaining the Ce^{3+} ion concentration constant in 3%Wt while the Cu^{2+} ion concentration was varied in 1, 2, 3, 10 and 20%Wt. After its synthesis, all the samples were annealed at 900°C by 24 h. The ZnO, $\text{ZnO}/\text{Ce}^{3+}$ and $\text{ZnO}/\text{Cu}^{2+}$ + Ce^{3+} powders were structurally characterized using X-ray diffraction (XRD) technique, and the XRD patterns showed that for pure ZnO, Cu^{2+} undoped $\text{ZnO}/\text{Ce}^{3+}$ and $\text{ZnO}/\text{Ce}^{3+}$ doped with the Cu^{2+} ion, the three samples exhibited the hexagonal wurtzite ZnO crystalline structure. However, the morphology and particle size of both samples were observed by means of a scanning electron microscopy (SEM); from SEM image, it is observed that the crystallites of both samples are agglomerated forming bigger amorphous particles with an approximate average size of 1 μm . In addition, the photoluminescence of the ZnO, Ce^{3+} doped ZnO and Cu^{2+} + Ce^{3+} doped ZnO samples was measurement under an illumination of 209 nm wavelength (UV region): for the $\text{ZnO}/\text{Ce}^{3+}$ sample, your emission spectrum is in the visible region from blue color until red color; the UV band of the ZnO is suppressed. The multicolor emission visible is attributed to the Ce^{3+} ion photoluminescence, while for the $\text{ZnO}/\text{Cu}^{2+}$ + Ce^{3+} , its emission PL spectrum is quenching by the Cu^{2+} ion, present in the ZnO crystalline.

Keywords

Zinc Oxide, Copper-Cerium, Co-Doped, Solution-Combustion

1. Introduction

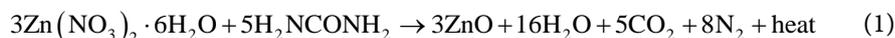
Between the II-VI semiconductors compounds, the Zinc oxide (ZnO) by its extraordi-

nary properties such as exhibiting an high energy band gap of 3.37 eV and an exciton binding energy of 60 meV [1] [2], has been converted in a strategic material for basic science investigation and technological applications [3] [4] [5], such as in the fabrication of solar cells [6], electro-optical devices [7], gas sensors [8], catalyst [9], piezoelectric device [10], paramagnetic materials [11] [12] [13], etc. In the scientific literature, there are various synthesis methods to obtain undoped and doped ZnO with various dopants types, for example: rare earth, metals, lanthanides, etc. [14], such synthesis methods include: electrodeposition [15], evaporation [16], vapor-liquid-solid (VLS) growth [17], metal organic catalyst assisted vapor-phase epitaxy [18], aqueous thermal decomposition [19], microwave activated chemical bath deposition (MW-CBD) [20], chemical bath deposition (CBD) [21], surfactant-assisted hydrothermal method [22], solution combustion [23] etc. Of the before methods, solution combustion presents some advantages because it is very fast, less expensive; it has an easier composition control, and coating can be deposited on large area etc. Lanthanide doped ZnO semiconductor nanoparticles have important technological properties: they have been used to make efficient photoluminescent materials when lanthanide ions are intercalated into ZnO crystal lattice, by means of an energy transfer process from the ZnO lattice to the Ce^{3+} ions to produce internal transitions atomic in the lanthanide ions. Between the lanthanide ions family members, the cerium ion (Ce) is the most emblematic according to the studies realized by various researchers [24] [25] [26] [27], from its results the authors show that the Ce^{3+} doped ZnO can present anomalous emission from the UV until red color region; this multiple emission is studied in the present work. As a complement for the present work, it is necessary to mention that the doping of the ZnO matrix with Cu^{2+} ions leads to sum or produce changes that improve the ZnO properties such as: band gap tailored, magnetic, electrical and optical. It can also produce passivation of defects and surfaces, as p-type dopant in the original n-type ZnO semiconductor. However, actually it is well known that due to the co-doping between the Cu^{2+} and Eu^{3+} ions in co-doped glasses matrix, the Cu^{2+} ion can quench the Eu^{3+} photoluminescence [28] [29] [30] [31], recently. S. Lopez-Romero *et al.* experimentally demonstrated that such quenching also can occur in solid ZnO semiconductor, this quenching effect can be used to tune between ultra-violet and visible PL emission in devices photoluminescent. In this study, ZnO/ Ce^{3+} and co-doped ZnO/ $Cu^{2+} + Ce^{3+}$ solid solutions powders have been synthesized by solution combustion method as a function of the Cu^{2+} ion concentration in %Wt. maintaining the Ce^{3+} ion concentration constant at a value of 3%Wt. for both compounds Cu^{2+} undoped and doped. Finally the samples were annealing at 900 °C by 24 h. In addition, the influence of the Cu^{2+} ion on the morphology, structure and photoluminescence of the compounds is analyzed.

2. Experimental Details

The experimental method of synthesis solution combustion basically consist in carry out a oxidation-reduction (REDOX) chemical reaction between an oxidizer agent and a fuel, such reaction is highly exothermic (1200 °C) [22] and generally to produce oxides

metallic, water vapor, carbon dioxide and nitrogen molecular. We used zinc nitrate hexahydrate ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) as oxidizer agent and urea (NH_2CONH_2) as fuel, the REDOX reaction between them is:



Complementary information about how the Equation (1) was obtained can be seen in reference [22]. Using the Equation (1) intrinsic ZnO, Ce^{3+} doped ZnO and $\text{Cu}^{2+} + \text{Ce}^{3+}$ co-doped ZnO were synthesized and after annealing at 900°C by 24 h, the source of the dopants Cu^{2+} and Ce^{3+} were CuCl_2 and Ce_2O_3 . The Ce^{3+} ion concentration was fixed at 3%Wt, while the Cu^{2+} ion concentration takes the values of 0, 1, 2, 3, 10 and 20%Wt. The ZnO/ Ce^{3+} and ZnO/ $\text{Cu}^{2+} + \text{Ce}^{3+}$ samples thus obtained were structurally characterized by x-ray diffraction (XRD) technique using a Philips PW 1800 diffractometer with $\text{CuK}\alpha$ radiation (1.5406 \AA), the morphology of the samples was recorded using a scanning electron microscopy SEM JEOL JSM 840.), the properties photoluminescent (PL) of the ZnO/ Ce^{3+} and ZnO/ $\text{Cu}^{2+} + \text{Ce}^{3+}$ samples was studied by means of a spectrofluorometer FluoroMax-P that uses a xenon lamp as excitation source, the wavelength excitation was of 209 nm.

3. Results and Discussion

3.1. Structural Characterization

Figure 1 shows the XRD patterns of the as prepared samples of ZnO, ZnO/ Ce^{3+} and ZnO/ $\text{Cu}^{2+} + \text{Ce}^{3+}$ as a function of the Cu^{2+} ion concentration with the Ce^{3+} ion concentration fixed in 3%wt. From the diffractograms can be seen that all the diffraction peaks can be indexed to the hexagonal wurtzite ZnO structure (JCPDS CARD #89-(102)), no change of the peaks toward lesser angles was observed, however in **Figure 2** can be observed the XRD pattern of the ZnO joint to two diffraction peaks at 28.4° and 32.5° : the first is assigned to the Ce_2O_3 phase and the second to the CuO compound. Also is observed that the diffraction peaks are very sharp indicating good crystallization of the products.

3.2. Morphology Study

Figures 3(a)-(d) are the SEM images of the ZnO/ $\text{Cu}^{2+} + \text{Ce}^{3+}$ samples with Cu^{2+} ion concentration of 0, 3, 10 and 20%wt respectively, it is observed that for low Cu^{2+} ion concentration the ZnO nanocrystals have nanoplate form and are agglutinated in amorphous geometry with bigger sizes of about $0.75 \mu\text{m}$ side and $1 \mu\text{m}$ large, while in the sample doped with 20%Wt the nanocrystals present ovoid form and are agglomerated in particles measuring up to $2.5 \mu\text{m}$ side and $3 \mu\text{m}$ large.

3.3. Photoluminescence Study

3.3.1. ZnO and ZnO/ Ce^{3+} Photoluminescence

Figure 4(a) and **Figure 4(b)** show the photoluminescence PL spectra of the pristine ZnO and of the Ce^{3+} doped ZnO nanocrystals respectively excited by a wavelength

radiation of 209 nm at room temperature. The PL spectra exhibited by the ZnO is typical of the pure ZnO: present a strong UV band peaking about 395 nm and a little broad green band centered about 460 nm. The UV emission band is due to excitonic recombination corresponding to the near band edge emission of the ZnO. The green emission band is attributed to deep-level or trap-state emission. Effectively, actually various mechanism have been proposed to understand the origin of the visible light of the ZnO which involve oxygen vacancies with various oxidation state: the single ionized oxygen vacancy, and the doubly ionized oxygen vacancy. Finally Vanheusened *et al.* [31] found that only the singly ionized oxygen vacancies can emit green light in the ZnO. For the Cu^{2+} undoped ZnO/ Ce^{3+} samples doped with 3%wt (see **Figure 4(b)**) and illuminated

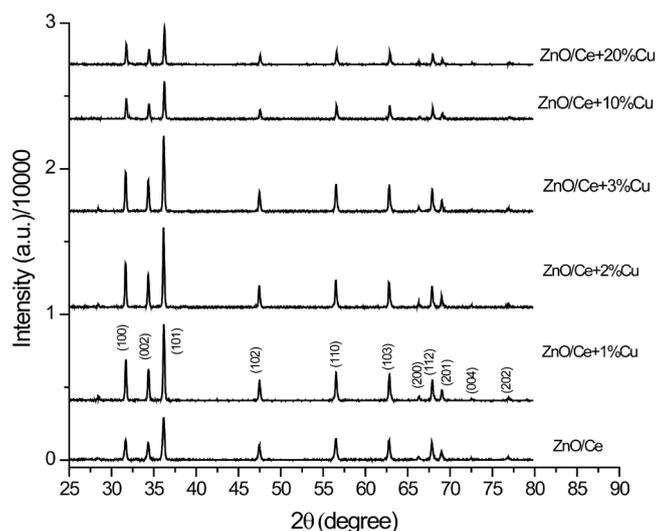


Figure 1. XRD pattern of ZnO, Ce^{3+} doped ZnO and $\text{Cu}^{2+} + \text{Ce}^{3+}$ co-doped ZnO as a function of the Cu^{2+} ion concentration.

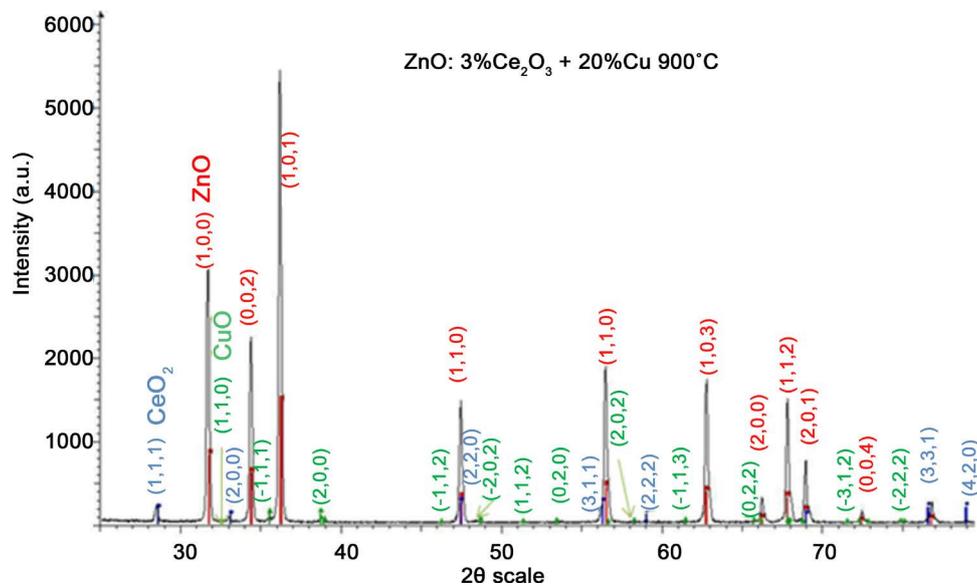


Figure 2. XRD patterns of ZnO, Ce_2O_3 and CuO compounds.

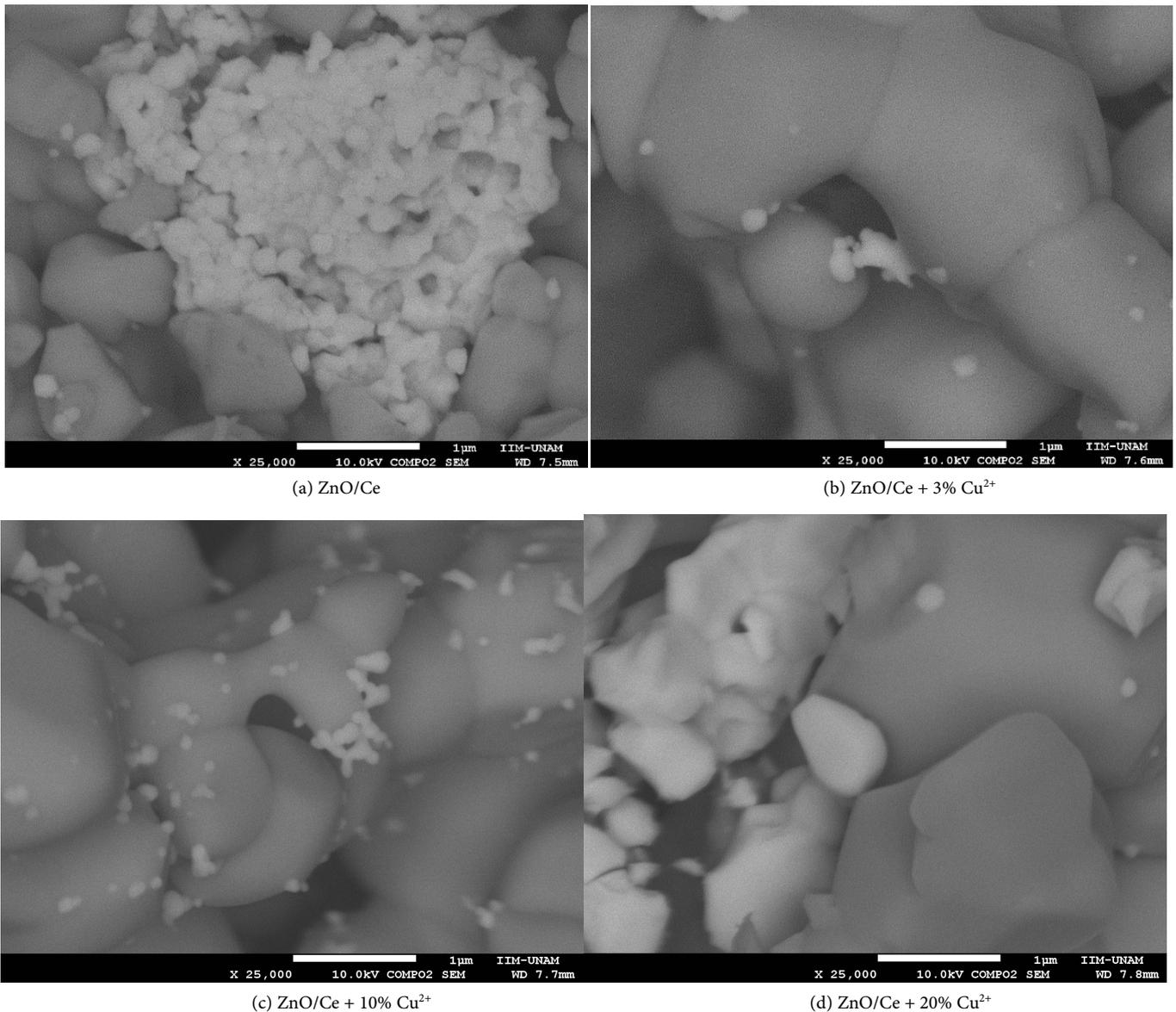


Figure 3. SEM images of the (a) Ce^{3+} doped ZnO, (b) Ce^{3+} co-doped ZnO with 3%Wt of Cu^{2+} , (c) $\text{Cu}^{2+} + \text{Ce}^{3+}$ co-doped ZnO with 10% Cu^{2+} , (d) $\text{Cu}^{2+} + \text{Ce}^{3+}$ co-doped ZnO with 20% Cu^{2+} .

with 209 nm wavelength. The emission spectra now present bands at 418 nm and 432 nm belong to the blue color, the peak centered about 452 nm correspond to purple color, the bands centered at 468 nm and 480 nm belong to the green region, the peak at 496 is green color, the band centered at 541 nm is yellow color, the peak at 648 nm is orange color, and the band at 687 nm is red color. This multicolor PL emission spectra is possible because when any matrix is doped with the Ce ion this attain two oxidation states namely the Ce^{3+} and Ce^{4+} , consequently f-d transitions occur by effects field in the ZnO matrix and 5 d state of the Ce^{3+} is splitting in two states: E_g and T_{2g} attributed to the new intern electronic ambient of the Ce^{3+} ion in the ZnO matrix [23]. It is important note the quenching of the UV band emission of the ZnO at 395 nm sowed by

the pure ZnO sample. This quenching effect is caused by defects on the ZnO surface generated by organic residues of the fuel (more studies are necessary with this tem).

3.3.2. Ce³⁺ Photoluminescence Quenching by the Cu²⁺ Ion

Figure 5 represent the excitation spectrum applied to a ZnO/Cu²⁺ + Ce³⁺ sample doped with 1%Wt of the Cu²⁺ ion, from the wavelength scale of the spectrum can be see clearly that for the excitation wavelength of 209 nm no PL emission of the ZnO/Ce³⁺ + Cu²⁺ sample was observed, only until a excitation wavelength of 378nm can be see a new emission spectrum of the Ce⁴³⁺ ion. The suppressing of the Ce³⁺ is due to that the Cu²⁺ ion quenching the Ce³⁺ photoluminescence [28] [29] [30]. This same quenching effect also occur in glass matrix doped with the europium and copper ions [29]: the authors

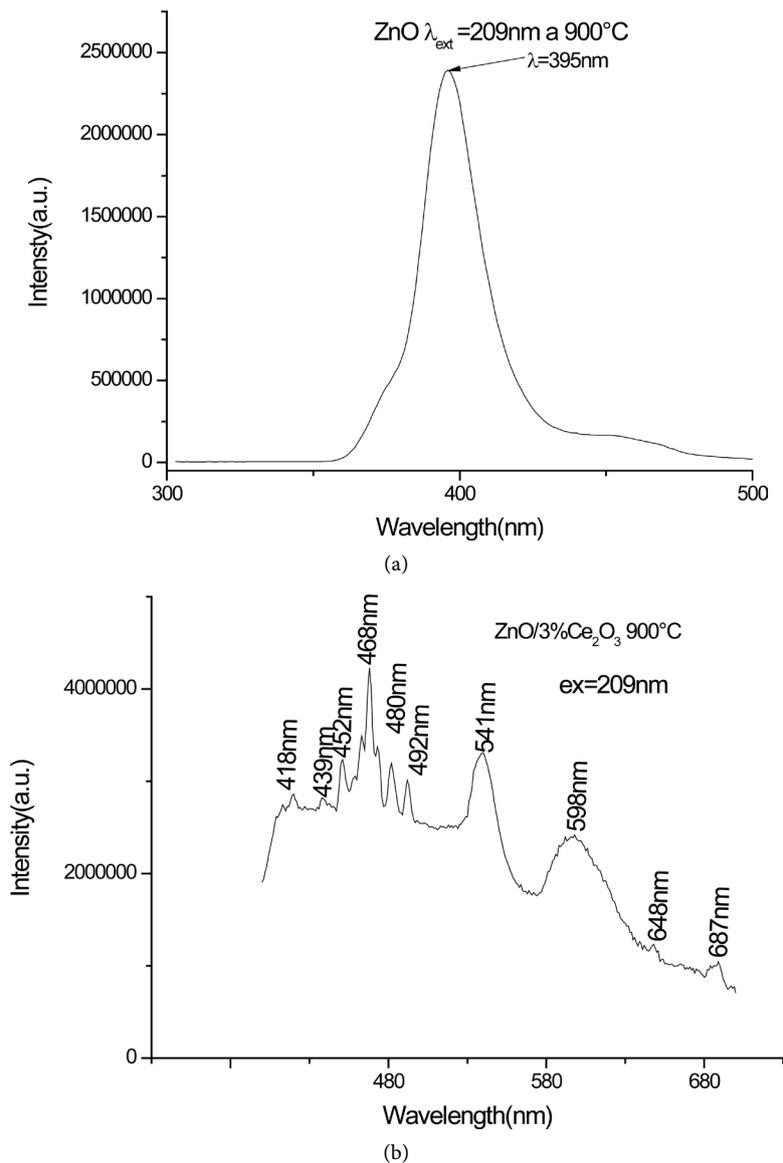


Figure 4. (a) Photoluminescence spectra of pure ZnO; (b) Photoluminescence spectra of Ce³⁺ doped ZnO. Doped with 3% of Ce³⁺ concentration.

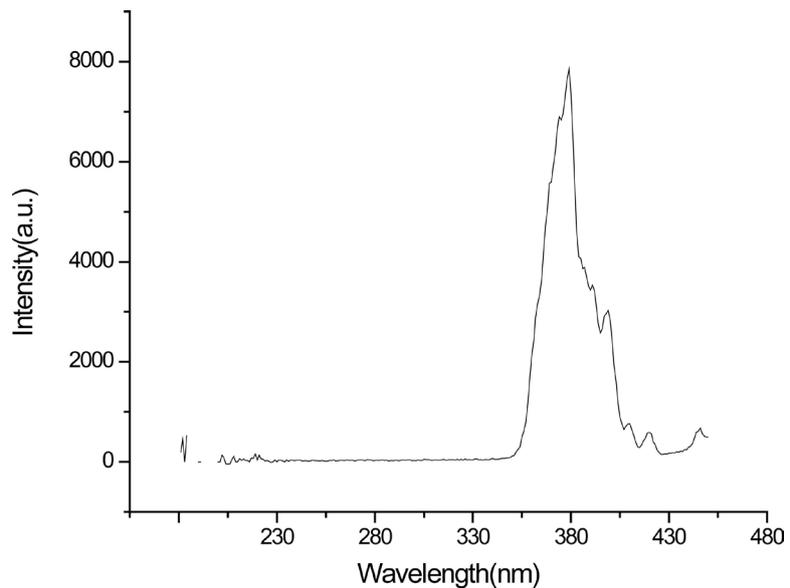


Figure 5. Excitation spectrum used to illuminate the ZnO/Cu²⁺ + Ce³⁺ sample doped with 1%Wt. of the Cu²⁺ ion.

explain the quenching effect by the occurrence of an overlap between Eu³⁺ emission band and a Cu²⁺ absorption band. In our case, we suppose that this quenching effect is due to an overlap between Ce³⁺ emission band and a Cu²⁺ absorption band. The Ce³⁺ PL quenching occur for all Cu²⁺ ion concentration. (More studies are necessary in this tem).

4. Conclusion

In this work, ZnO, Ce³⁺ doped ZnO and Ce³⁺ + Cu²⁺ co-doped ZnO were synthesized by a solution combustion method maintaining the Ce³⁺ ion concentration constant and changing the Cu²⁺ ion concentration. The ZnO resultant crystalline structure was the hexagonal wurtzite structure for the three samples types. From PL studies, it is found that the UV band of 395 nm in the pure ZnO is suppressed in the Ce³⁺ doped ZnO and the PL emission spectrum of the Ce³⁺ + Cu²⁺ co-doped ZnO samples also was quenching by the Cu²⁺ ion.

Acknowledgements

The authors wish to thank to Dr. Ciro Falcony (IPN), Adriana Tejeda (IIM) for the XRD measurements, to Omar Novelo Peralta (IIM) for his SEM study, to M.A. Canseco Martinez and Lázaro Huerta (IIM) for their chemical analysis.

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