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Preparation and Characterization of the Thermoluminescence Properties of Mg-doped ZnO

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Abstract

ZnO and Mg-doped ZnO nanoparticles were prepared by sol-gel method. X-ray diffraction studies of the sample confirm hexagonal phase. Crystallite size calculated using Scherer equation and found to be 48.5nm for undoped ZnO and 44.3nm for 1% Mg doped ZnO, 40.9nm for 2% Mg-doped ZnO and 38.7nm for 3% Mg-doped ZnO. Undoped sample exhibit unstructured low intense TL glow curve, whereas Mg doped samples exhibit will structured TL glow curves with a broad peak around 280^0C to 330^0C depends on the UV energy. The TL response as a function of UV energy showed a linearity behavior over the range investigated, and 1% Mg-doped ZnO sample gives very good reproducibility compared with 2% and 3% Mg-doped ZnO samples.

Keywords: Mg-doped ZnO; sol-gel method; UV source; TL.

1. Introduction

Zinc oxide is a chemical compound found naturally in the mineral called zincite and has a wide band gap semiconducting material with several applications due to its unique physical and chemical properties [1,2]. ZnO crystallites consist of a hexagonal wurtzite lattice with lattice parameters a = 3.296 A and c = 5.206 A , recently ZnO has attracted substantial interest because of their use in many applications such as in light-emitting diode [3,4] , solar cells [5], UV light detectors [6], and field emission devices [7].

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Many chemical methods have been applied to synthesize ZnO crystalline, i.e., precipitation [8], Sol- gel [9], solution combustion, thermal decomposition [10] and hydrothermal [11]. The sol-gel method has attracted attention as it is a relatively low cost process and represents an ideal method for growing ZnO nanostructures for use in many applications such as biomedical devices [12]. Doping of impurities provide a way to modify the properties of nanostructures by changing their compositions [13], ZnO doped with Mg permits band gap narrowing [13,14] and wavelength tenability [15] to blue or even green light spectra range by creating barrier layers, which will facilitate radiative recombination by carrier confinement [16,17]. Thermoluminescence "TL" as mentioned by McKever and his colleagues is one of the processes in thermally stimulated phenomen. In general view, thermoluminescence is a temperature stimulated light emission from crystal after removal of excitation. TL technique is used to measure the radiation dose absorbed by material. Many phosphor materials, synthetic as well as natural, have been characterized to evaluated its feasibility as thermoluminescence dosimeters "TLD" applicable in several dosimetry fields, including low –dose and high- dose applications such as environmental, clinical, personal, industrial. ZnO TL properties were firstly reported in the late 60’s and early 70’s, concerning on low temperature TL features as a tool to study defects, with no interest in dosimetry applications [18,19]. Cruz and his colleagues reported on the synthesis of ZnO nanophosphors by thermal annealing of ZnS synthesized using a chemical bath deposition reaction, with TL properties suitable for use in TL dosimetry [20]. After that many authors reported on the TL properties of both ZnO and doped ZnO, synthesized through different chemical methods in an attempt to improve their dosimetric characteristics [21-24]. The aim of this work is to study the TL properties of Mg-doped ZnO nanoparticles prepared by Sol-gel method.

2. Experimental procedure

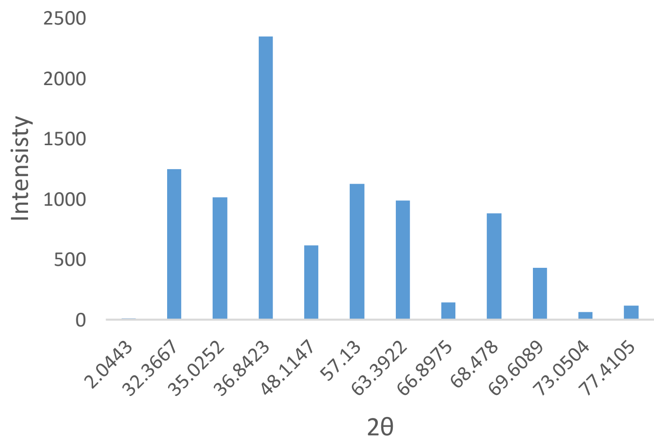
Pure and Mg doped ZnO nanoparticles are prepared by sol – gel method. The starting materials are zinc nitrate, magnesium nitrate, acetic acid and EDTA. The experiment is performed by dissolving stoichiometric amount of above reagents together in minimum amount of distilled water. ZnO nanoparticles are prepared with three concentrations (1%,2%,3%) the obtained powder is subjected to calcinations at 600 ºC for 2hours, then 0.1gram of powder was placed in a 0.85cm diameter cylindrical mold for compression to about 1 ton using a hydraulic press to make pellet shaped sample.

3. Characterization

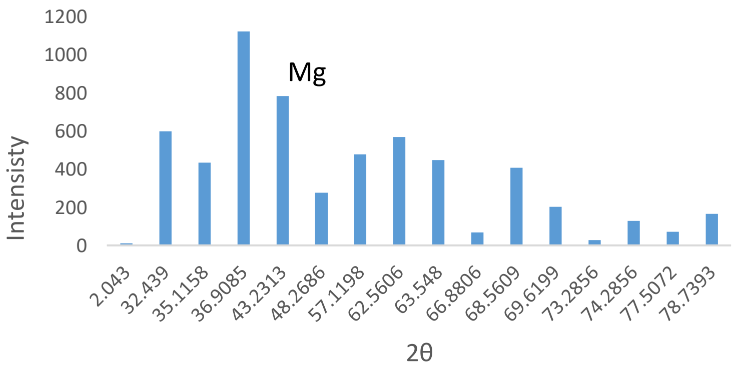
The structural properties of nanoparticles determined from X-ray diffraction (XRD) spectra by diffractometer (PW1800) equipped with Cu kα radiation ($\lambda = 0.154$nm) with scan rate of a 5^0/sec. FTIR spectra were recorded using IR – Affinity-1 in the range of 400 to 4000cm^{-1}. The Mg-doped ZnO TL yield was readout by a Harshaw 3500 TL reader located at sebha university , Libya. In order to eliminate the contribution by triboluminescence all readings were performed in an atmosphere of high purity N_2 preheat temperature is 160 ºC for 10s; readout temperature of 400 ºC for 13.33s and heating rate cycle of 25 ºC s^{-1}. Then, annealing temperature of 400 ºC was applied for 1h to erase any thermoluminescence signal, and the irradiation source was a UV lamp.

4. Results and discussion

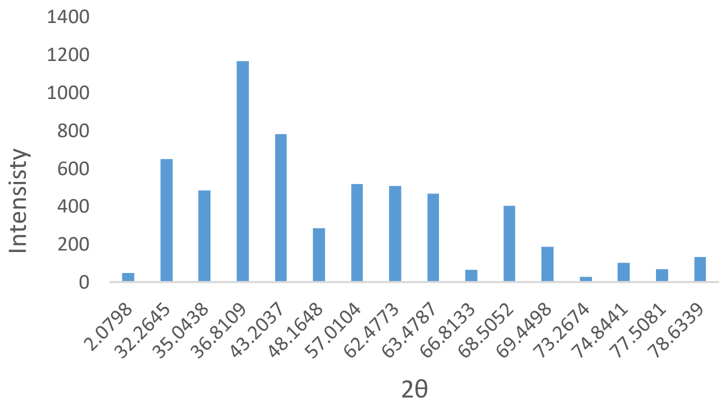
Figs (1-4) shows XRD diffraction pattern of ZnO and Mg-doped ZnO nanoparticles. The diffraction peaks showed the crystalline with peaks corresponding to (100), (002), (101), (102), (110), (103), (200), (112), and (201) planes the diffraction patterns of samples a,b,c, and d are consistent with the reference codes 01-075-0576 and 03-065-0523. The peak at plane (101) positioned at 36.79^0 for sample a, and at 36.9^0 for sample b, at 36.31^0 for sample c, and at 36.83^0 for sample d, which has maximum intensity. The obtained XRD spectra showed extra peak may corresponding to Mg^{2+} appears at 43.20^0, the changes in cell parameters and peak positions shift towards higher 2θ value was observed in XRD data, which indicating successful substitution of Mg^{2+} in host ZnO nanoparticles and also due to the smaller ionic radius of Mg^{2+} compared to Zn^{2+} [25,26].



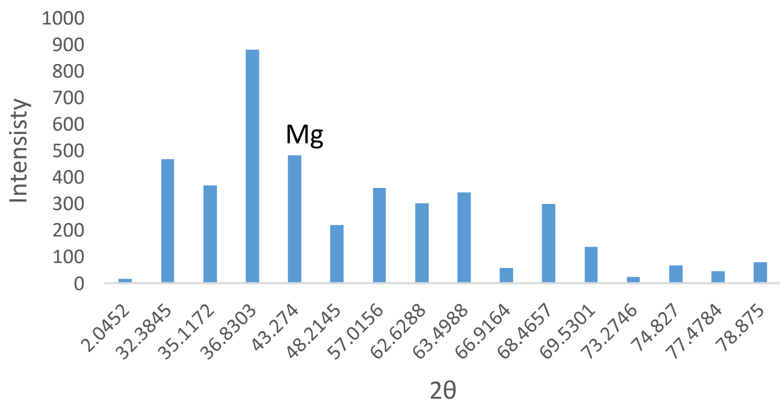
0% Mg-doped ZnO



1% Mg-doped ZnO



2% Mg-doped ZnO



3% Mg-doped ZnO

Figures 1- 4: XRD pattern of ZnO and Mg-doped ZnO

Crystallite size of ZnO and Mg-doped ZnO was calculated by Debye-Scherer formula [27].

$$D = \frac{k\lambda}{\beta \cos\theta} \rightarrow (1)$$

Where D is the crystal size, k is a constant which equal 0.89 for the hexagonal structure, λ is the wavelength of X-ray used (1.5418 A^0), β is full width at half maximum and θ is the Bragg's angle. The calculated average crystal size was found to be 48.5nm for ZnO , 44.3nm for 1% of Mg-doped ZnO, 40.9nm for 2% Mg-doped ZnO, and 38.74nm for 3% Mg-doped ZnO, which is decreases by increasing the concentration of Mg as shown in Fig (5). The crystal size decreases with Mg content which is may due to the substitution of Mg metal into zinc oxide lattice. Also lattice parameters are calculated and the results are shown in table (1).

Table 1: shows Lattice parameters for ZnO and Mg-doped ZnO nanoparticles.

ZnO/ZnO:Mg	a(nm)	C(nm)	c/a
0 %	0.3196	0.5127	1.6
1%	0.31843	0.5111	1.6
2%	0.3201	0.5121	1.599
3%	0.3208	0.5110	1.592

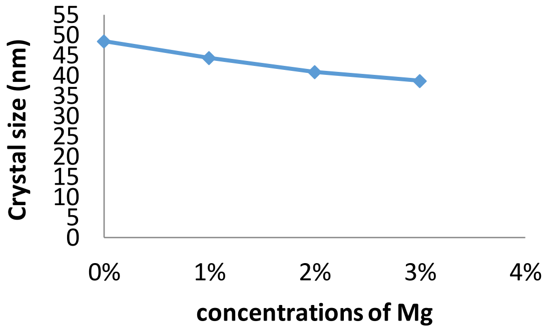
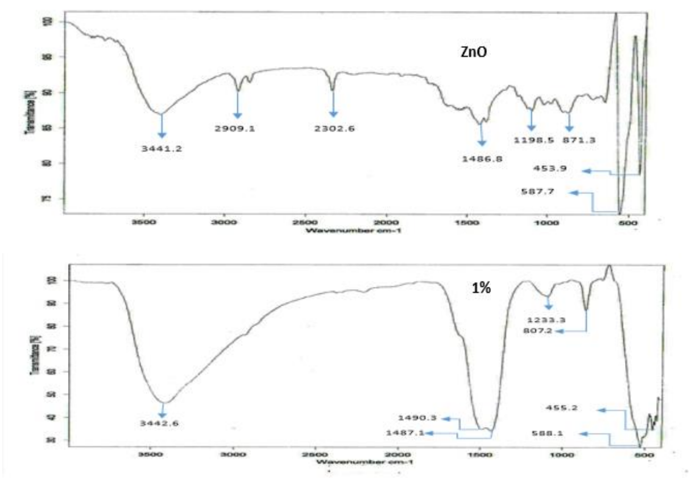


Figure 5: Crystal size versus Mg concentration

5. FT-IR

This technique used to obtain information regarding chemical bonding in a material, the band positions and number of absorption peaks depends on crystalline structure, chemical composition and morphology. Fig (6a,6b,6c,6d) shows the IR spectrum of ZnO and Mg-doped ZnO nanoparticles recorded in the range of 400 to 4000cm^{-1}. The KBr technique has been used to record the spectra, the absorption band near 440 cm^{-1} which correspond to Zn-O peak, the absorption band at around 3450cm^{-1} corresponded to the O-H group stretching vibration of water presence in ZnO, the peak at 1520cm^{-1} indicating the H-O-H bending vibrations. The peaks around 2350cm^{-1}are due to the presence of carbon dioxide, the absorption peak around 1400cm^{-1} is assigned to the bending vibration of C-H stretching.



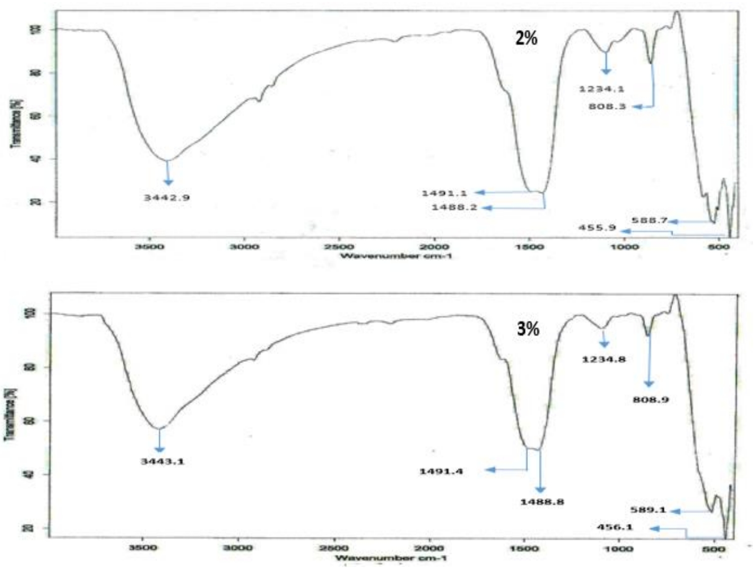


Figure 6: The FTIR pattern of ZnO and Mg –doped samples.

6. Glow curve

Figure (7) shows the glow curves as obtained from Mg-doped ZnO samples when exposed to UV radiation. The TL intensity increased as exposure time increase, also the TL peaks shift to higher temperature when the exposure time increases. The pure ZnO samples are very sensitive to visible light compared to Mg-doped ZnO. The shape of the glow curves for Mg-doped ZnO are simple and the TL peak with maximum intensity located between 280 ^{0}C and 330 ^{0}C. The increase in intensity of TL glow peak with exposure time and energy is due to more and more traps responsible for the glow peak are getting filled with the increase of exposure time and energy and subsequently these traps release the charge carriers on thermal stimulation to finally recombine with their counterparts, which giving rise to glow peaks. Cruz and his colleagues [28] they found that the TL peaks with maximum located at ~200 ^{0}C and 250 ^{0}C when irradiated by beta particles, and N. Pushpa and his colleagues [29] also found well-structured TL peaks at ~345 ^{0}C. It seems that Mg2+ as dopant help in generating more number of electron / hole traps and luminesce centers which are responsible for TL variation of the glow peak intensity.

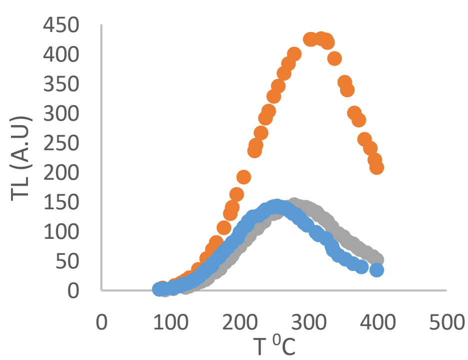


Figure7 : TL glow curves of Mg-doped ZnO samples irradiated with UV – Source for 5min – 10 min – 20min .

7. Reproducibility

In order to study the reproducibility of the material at certain doses, a total of four Mg-doped ZnO samples were used, The test was performed for ten consecutive cycles, thermal annealing treatment, irradiation and reading. Irradiation was performed at 600µJ/cm^2 UV for 5 min. The reproducibility of the TL response as a function of absorbed dose was calculated by equation (1).

$$R = \frac{100\sigma}{\overline{X}} \leq 7.5\% \rightarrow\rightarrow (2)$$

Where σ is the standard deviation and $\overline{X}$ is the average of all readings during the 10 cycles. The typical reproducibility values are between 2% and 10% [30,31,32]. The results as shown in fig(8) that 1% Mg-doped ZnO has very good performance, with reproducibility value below 3% then 3% Mg-doped ZnO which has reproducibility value equal to 4.6% and 2% Mg-doped ZnO the reproducibility value is 12.7%.

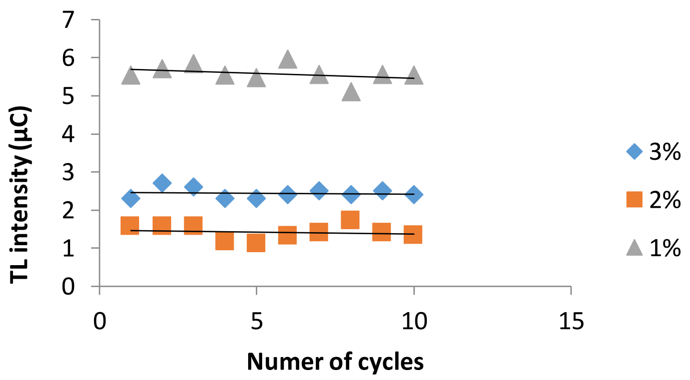


Figure 8: Reproducibility test for Mg- doped ZnO.

8. TL response

One of the important characteristic to be a good thermoluminescence dosimeter is linear relationship between TL emission and the absorbed dose. The particular thermoluminescence material gives the great effect to the linearity range. Generally, the response of TL phosphors is linear at low absorbed dose value than becomes supralinear and finally saturates at high values [33]. The Mg- doped ZnO with various concentrations were exposed to UV energy. Figure (9) shows a linear TL response from 100µJ/cm^2 up to 600µJ/cm^2, the regression (R^2) start from 0.93 to 0.98 for the three concentrations as shown in figure (9).

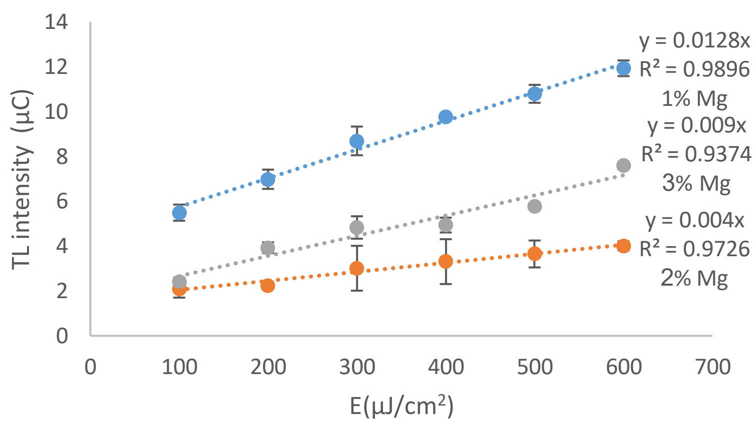


Figure 9: shows the linearity of Mg-doped ZnO samples when exposed to UV radiation

9. Sensitivity

TL sensitivity can be define as the amount of light released by phosphor per unit of radiation exposure [34]. There are many factors that affect to TL sensitivity dosimeter such as kind and concentration of dopant, system of the readout and heating rate. The least square fit of the change in TL response for 1% Mg-doped ZnO to be 1.42 greater than that obtained for 3% Mg-doped ZnO and 3.2 greater that obtained for 2% Mg-doped ZnO irradiated at same UV energy. Therefore, the 1% Mg-doped ZnO has better response and is more sensitive compared to 2%, 3% Mg-doped ZnO.

10. Conclusion

The Mg-doped ZnO nanoparticles have been successfully synthesized by sol – gel method. The ZnO exhibits hexagonal wurtzite structure. Doping with Mg reduced the particle size from 48.5nm for undoped ZnO to 38.7 for 3% Mg-doped ZnO. TL study of UV radiated Mg-doped ZnO exhibit one peak located between 280 0C and 330 0C depends on energy and exposure time, and the TL intensity increase with increasing the energy and exposure time. 1% Mg-doped ZnO has very good reproducibility compared with 2% and 3% Mg-doped ZnO, and the TL sensitivity of 1% Mg-doped ZnO is greater than 2% and 3% Mg-doped ZnO. Hence, Mg-doped ZnO material have a good TL response and may be useful for environmental dosimetry.

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