J. Myanmar Acad. Arts Sci. 2020 **Vol. XVIII.No.2B**

STRUCTURAL CHARACTERIZATION AND NON-OHMIC PROPERTIES OF AL DOPED ZNO VARISTORS

Win WinYee[1], Pwint Hlaing Ni[2], Zin Oo Hlaing[3], May Aye Khine[4] and PwintYi Thein[5]

Abstract

Al (0 mol%,1 mol% & 2 mol%) doped ZnO ceramics are prepared by using solid state reaction method. Samples are heat treated at 500°C for 3 hrs. After pre-heat treatment schedule, samples are heated treatment again temperature 1100°C for 2 hrs respectively. Structural characterization of the samples are investigation by using XRD. From the XRD analysis, structural properties such as, lattice parameters, crystallite size and micro stain are evaluated. Energy gaps of the samples are studied by using UV Vis spectrometer. Non-Ohmic behavior of the Al doped ZnO ceramics varistors are also determined.

Keywords: XRD,ceramics & varistor.

Introduction

Varistors are used to protect a circuit from high voltage surges. When a voltage surge is applied to s circuit, the outcome is usually catastrophic to the circuit. A capacitor may be installed across the signal lines. However, this capacitor cannot suppress voltage surge. Therefore, when circuit protection from voltage surge is required, a varistor is used as a voltage protection device. When a voltage surge exceeding a specified value (varistor voltage) is applied, the varistor suppresses the voltage to protect the circuit. When voltage surge does not exceed the varistor voltage, the varistor works as a capacitor.

The varistor is composed of ZnO with small addition of Bi, Co, Mn and other metal oxides. The structure of the body consists of a matrix of conductive ZnO grains separated by grain boundaries providing P - N junction semiconductor characteristics. These boundaries are responsible for blocking conduction at low voltages and are the source of the nonlinear conduction at higher voltages.

It is well known that, the performance of ZnO varistor is controlled by the dopant additives, usually metal oxides and the dopants are responsible for the formation of varistor behavior. It is believed that, dopants play an important role to modify the defect concentrations at the ZnO grain and /or of grain boundary where the performance of ZnO is sensitive to the some additives even when their amount is very small. Among the additives, Al is widely used as grain growth enhancer and ZnO conductivity enhancer in ZnO based ceramics.

In this research work, the introduction of Al doped ZnO ceramics samples were prepared by solid state sintering method. X - rays diffraction is used to characterize the structural properties of the ceramics samples. ln I – ln V characteristics of the samples are studied. From the I (V) data, varistor behavior of the samples are evaluated. Bandgaps of the prepared ceramics samples are examined by UV Vis spectrometer.

[1] Lecturer, Department of Physics, University of Computer Studies, Yangon.
[2] Lecturer, Department of Physics, University of Information Technology, Yangon.
[3] Dr, Lecturer, Department of Physics, University of Maubin.
[4] Dr, Associate Professor, Department of Physics, University of Kyaing Tong.
[5] Dr, Professor, Department of Physics, Nationalities Youth Resources Development Degree College, Yangon.

Experimental Procedure

Al doped ZnO samples were mixed, according to (1 - x) ZnO + (x) Al_2O_3, where, x = 0, 1 mol %, 2 mol % respectively. Analar grade ZnO and Al_2O_3 were mixed with agate mortar for 2 hrs. After mixing the specimen, the mixture was heat treated at 500^{o}C for 3 hrs. After that, the mixture was grinded with ball milling for 6 hrs. Then, the mixture was heat treated again at 1100^{o}C for 2 hrs.

Structural characteristics of the ceramics were investigated from the XRD spectra. Samples were scanned from 10^o to 70^o in diffraction angle, 2Θ, with scanning speed 0.01 degree/ sec. The diffraction patterns were recorded at room temperature, using CuK_α (λ = 1.5408 Ȧ) radiation, and voltage and current were 40 kV and 50 mA respectively.

Optical characteristics of the ceramic samples were examined in the wavelength 190 nm to 700 nm, using UV Vis spectrometer ,(SHIMADZU UV-1800). From the optical absorption spectra, band gaps of the samples were studied.

The mixture powder were uniaxially pressed into circular shape disc of 20 mm in diameter and 3.5 mm in thickness at a pressure of 19.5 tons. Silver paste was coated on the both surfaces of the samples and the electrodes were formed by heating at 600^{o}C for 15 min.The nonlinear behavior of the ceramic samples were measured using high voltage DC power supply. The threshold voltage (V_{1mA}) was measured at current 1mA, and the leakage current was studied at 0.8 V_{1mA}. From the lnV - lnI characteristics, nonlinear coefficient of the ceramics was examined.

Results and Discussion

XRD spectra of Al (0 mol %,1 mol % and 2 mol %) doped ZnO ceramic samples are depicted in figure (1). It is obvious that, only diffraction peaks of hexagonal wurtize ZnO (075 - 0576 > JPDCS library file) are observed, as seen in figure (1). Furthermore, (101) peak is the most intense peak and a little shift of (101) peak is found in all spectra. Atomic radius of Al atoms are slightly smaller than Zn atoms and the covalent bond length of Al - O is estimated to be shorter than Zn - O. Al has solid solubility limit of about 2~3 % in ZnO . These results can interpret as Al ions are possibly diluted in the ZnO host matrix. Lattice parameters ‘a’, ‘c’ and lattice tetragonality c/a are listed in table (1).Crystallize size and micro stain, derived from Debye-Sherrer equation , are also listed in table (1).The ‘a’ and ‘c’ parameters of the ceramic samples are very little change, as listed in table (1), it may be due to the unchanged of hexagonal wurtize ZnO structure.

Table 1 Structural properties of ZnO : Al ceramics.

Molar concentration	lattice parameter “a(A^o)”	lattice parameter “c(A^o)”	lattice distortion	crystallize size (nm)	micro strain	FWHM of (101) peak(^{o})
Pure ZnO	3.2484	5.2056	1.6027	86.173	2.6377x 10^{-3}	0.097
Al 1mol%	3.2497	5.2049	1.6017	55.370	1.2928 x 10^{-3}	0.151
Al 2mol%	3.2438	5.2031	1.6045	42.219	2.0100 x 10^{-3}	0.198

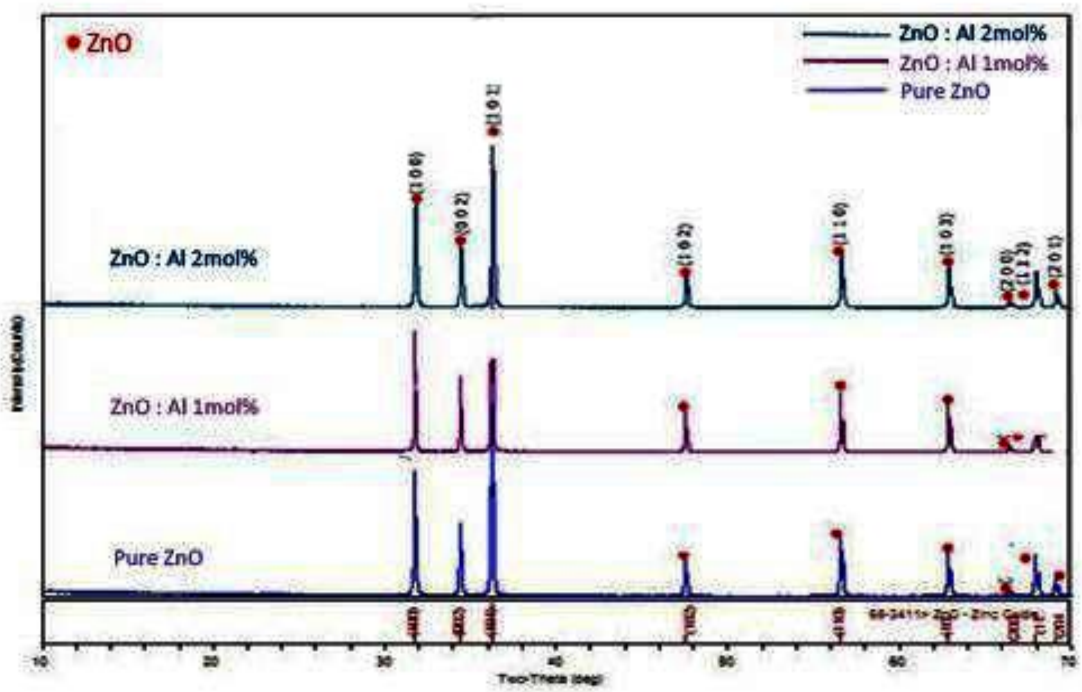

Figure 1 XRD spectra of Al doped ZnO ceramics with different Al contents.

Nonlinear properties of the ceramic samples are studied from the lnV vs lnI curves, as seen in figure (2). Nonlinear coefficients are obtained from the slopes of the ln V vs ln I curves. Threshold voltages (V_{1mA}) which are measured at current 1mA and leakage currents are examined at 0.8 V_{1mA}.Data are collected and listed in table (2). The ZnO grains themselves are highly conductive, while the intergranular boundary formed of other oxides is highly resistive. Only at those points where ZnO grains meet does sintering produce microvaristors, comparable to symmetrical back-to-back Si Zener diode. The nonlinear behaviour of the varistor results from the number of microvaristors connected in series or in parallel.

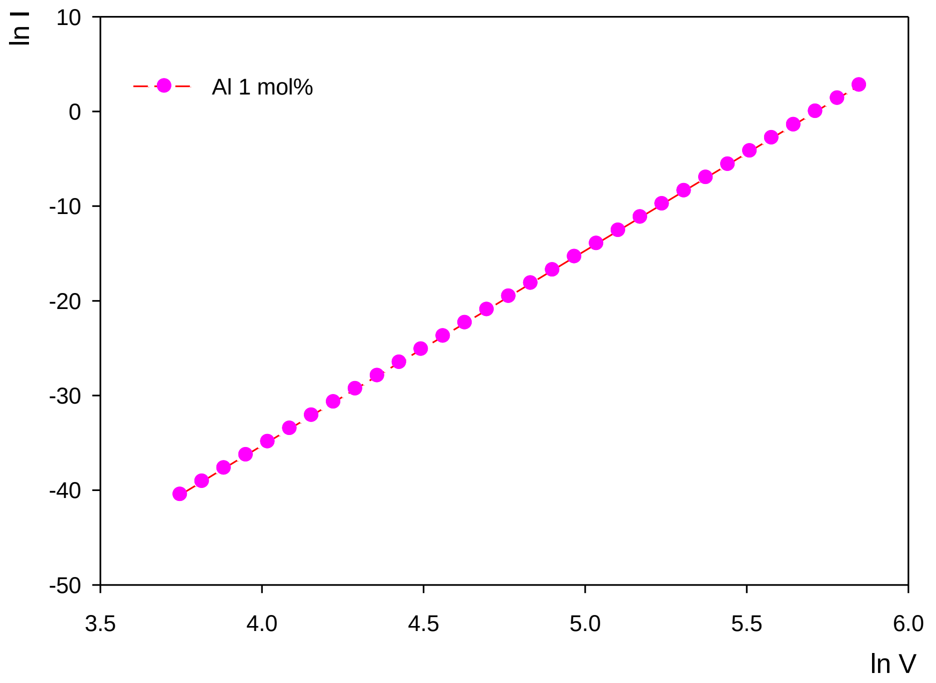

Figure 2 (a) Non- linear behavior of Al 1 mol % doped ZnO ceramics.

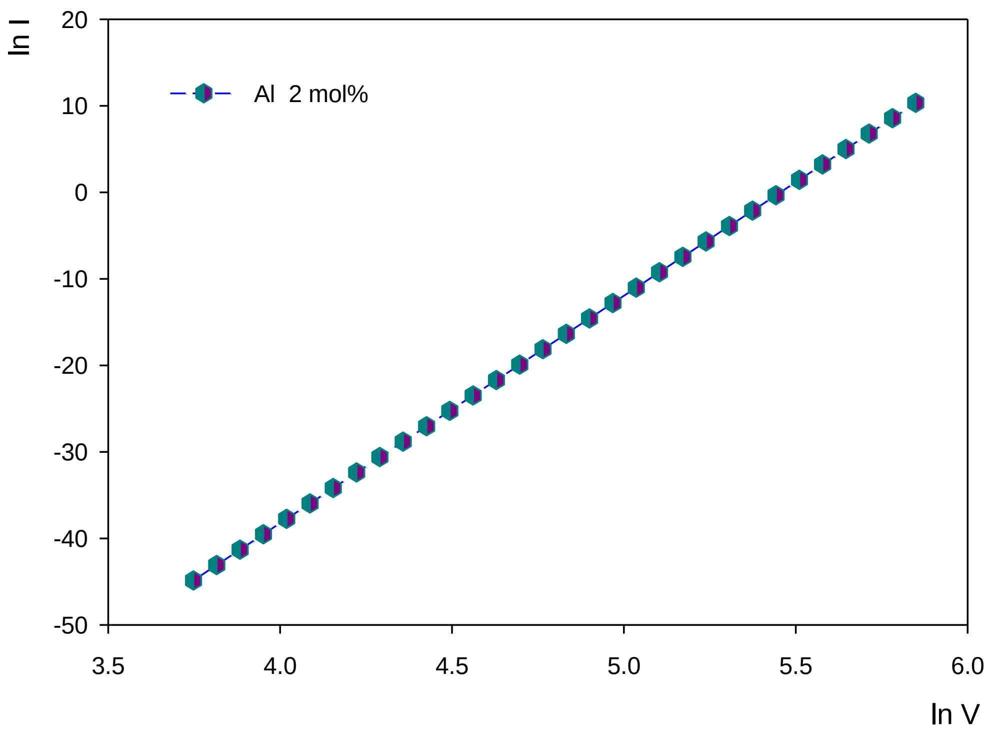

Figure 2 (b) Non- linear behavior of Al 2 mol % doped ZnO ceramics.

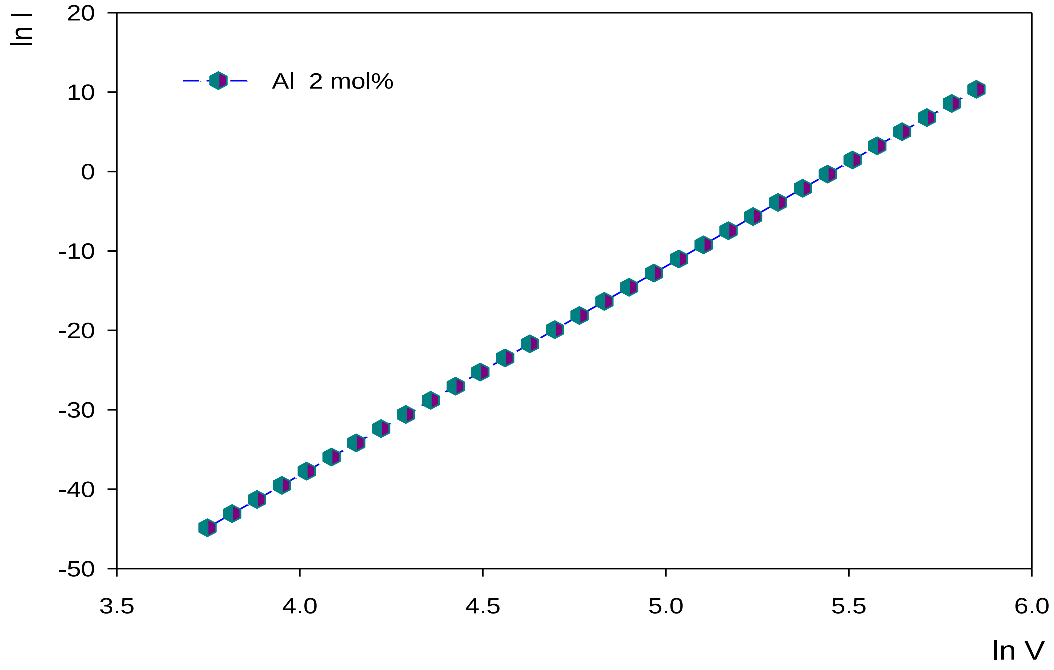

Figure 2 (c) Non- linear behavior of Al 2 mol % doped ZnO ceramics.

Table 2 Varistor Properties of ZnO : Al Ceramics.

Al concentration	**Th Threshold Voltage V_{th}(V)**	**Leakage Current I_L(mA)**	**Non-linear coefficient**
Al 1mol%	215.1	0.011219	20.15
Al 2mol%	213.4	0.011156	20.18

It is obvious that the nonlinear coefficient increases with increasing Al content, as listed in table (2). It is believed that after doping Al in to ZnO ceramics, there were be good grain boundaries in host ZnO ceramics. From the ln V vs ln I curves, nonlinear coefficients of the samples are obtained by the following relation.

$$\propto = \frac{\log\left({}^{I_2}/_{I_1}\right)}{\log\left({}^{V_2}/_{V_1}\right)}$$

where, I_1= 1 mA and I_2 = 10 mA and, V_1 and V_2 are the voltages corresponding to $I_1$and I_2Threshold voltages ($V_{1\ mA}$), which are measured at current 1 mA and leakage currents are studied at 0.8 $V_{1\ mA}$. Data are collected and listed in table (2).

Threshold voltage and leakage current decrease when Al content is raised, as listed in table (2).It may be due to increase of grain size that leads to lower the threshold voltage. The substitution of Zn^{2+} ions with Al^{3+} in ZnO lattice causes the increase of electrical conductivity, due to increase of charge carrier .This result can interpret as a decrease of leakage current.

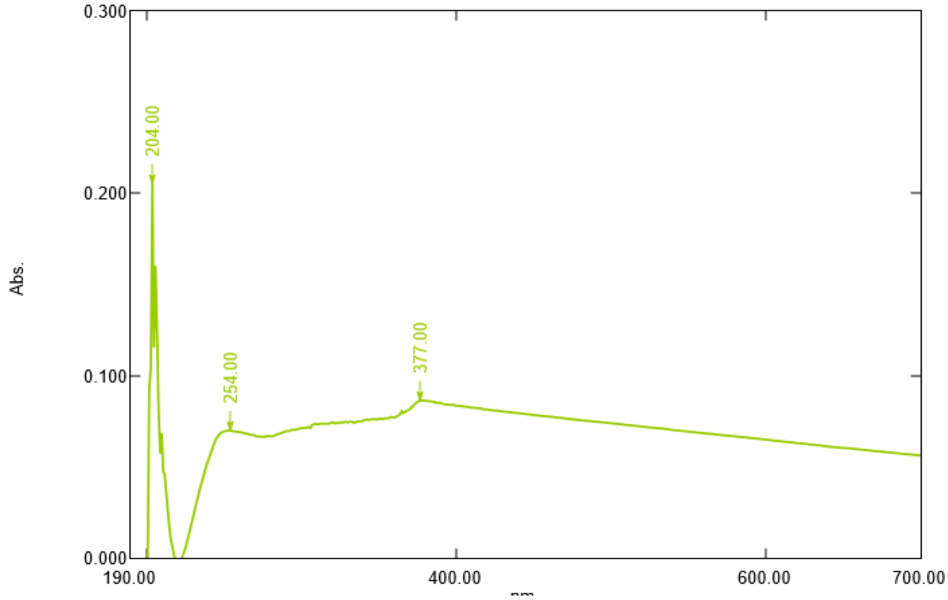

Figure 3 (a) Optical absorbance spectrum of pure ZnO

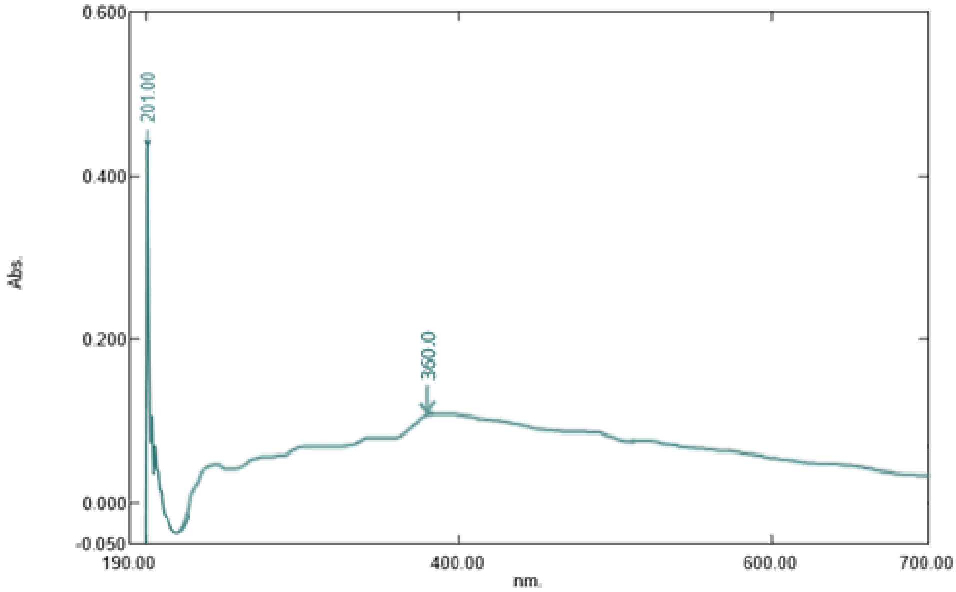

Figure 3 (b) Optical absorbance spectrum of Al 1 mol % doped ZnO ceramics

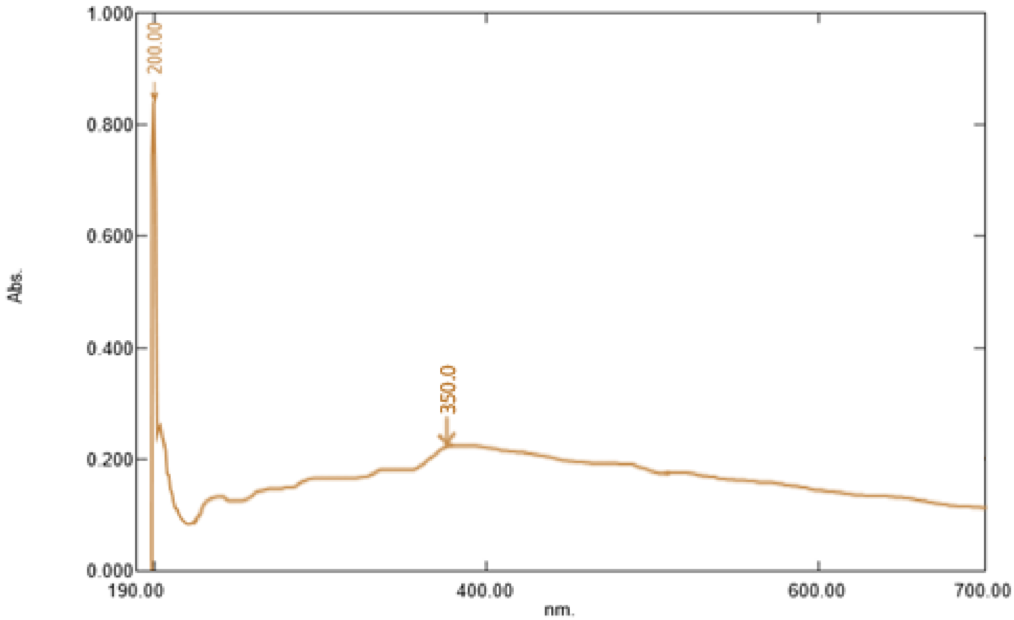

Figure 3 (c) Optical absorbance spectrum of Al 2 mol % doped ZnO ceramics

Figure (3) depicts the optical absorption spectra of ZnO : Al ceramics in the wavelength 190 nm to 700 nm. It is obvious that ,absorbance increases with Al content. The optical bandgaps of the ceramic samples were examined by applying Tauc - Mott relation and listed in table (3). It is noted that bandgap varies with dopant Al content. It is probably due to the quantum confinement effect in compound semiconductors.

Table 3 Band gaps of Al doped ZnO ceramics.

Al concentration	**Energy gap (eV)**
Pure ZnO	3.274
Al 1mol%	3.429
Al 2mol%	3.527

Conclusion

Al (1 mol % , 2 mol %) doped ZnO ceramic samples were prepared by applying solid state reaction method in this research. Structural properties of the samples were characterized by using XRD technique. Influence of Al concentration on lattice parameters, lattice distortion, crystallite size and lattice micro strain were determined. Ceramic samples were prepared with standard varistor preparation process and varistor behaviour of the samples were examined. From the V (I) data, threshold voltage, leakage current and nonlinearity factor were studied. Furthermore, optical bandgaps of the samples were evaluated.

Acknowledgements

The authors would like to acknowledge the Universities Research Centre (URC) , University of Yangon, for XRD measurements.

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