

FABRICATION AND CHARACTERIZATION OF $\text{Zn}_{1-x}\text{Cu}_x\text{O}$ CERAMICS COMPOUND

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Abstract

Copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) ceramic compound is a transparent conducting oxide. Copper zinc ceramic compound was prepared for mixing and grinding by high purity of ZnO (zinc oxide) powder and CuO (copper oxide) powder according to the stoichiometric composition. Copper zinc ceramic oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) ceramic compound was prepared at various growth temperatures by solid state method. The crystalline properties of copper zinc ceramic compound were investigated by X-ray diffraction (XRD). Copper zinc oxide ceramic compound were also studied by electrical properties.

Keywords- sample preparation, XRD result, conductivity and resistivity measurement

Introduction

Ceramics are classified as inorganic and nonmetallic materials that are essential to our daily lifestyle. Ceramics are favored for many sensors' application because of their wide availability, low cost, and ease fabrication (Yin, Q., Zhu, B., and Zeng, H.2010). The electrical properties of ceramics depend on composition, temperature, atmosphere and microstructure. Nanocrystalline metal oxide has numerous important properties like catalytic, electrical and optical properties. Hence ZnO is one of the metal oxides which attracts due to its band gap energy of 3.37 eV and a larger excitation binding energy of 60 meV at room temperature. ZnO was intensively studied as a material dilute magnetic semiconductor (Kittle C., 1986). When ZnO is doped with the transition metal ions such on Mn, Fe, Cu, Cr and Co, it shows ferromagnetic properties. The optical and electrical properties of ZnO must be tailored effectively to the desirable range by doping with suitable elements (Pillai S.O., 2005).

The Cu doping of ZnO material has been an active research area on the last few years, due to its deep acceptor level, enhanced green luminescence, gas sensitivity etc. The high ionization energy and low formation energy of Cu makes it a fast diffusing impurity into ZnO lattice. Depending upon the oxidation states of Zn, Cu doped ZnO becomes a p type semiconductor (D.B. Buchholz *et al.*, 2005). Ceramic materials are special because of their properties. They typically pass high melting points, low electrical and thermal conductivity values, and high compressive strengths. This paper reports the XRD study and copper zinc oxide ceramic compound were measured by conductivity (Buchanan, R. 1984).

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Material & Methods

Powder Sample Preparation

In the main system, 0.05 mole of CuO and 0.95 mole of ZnO were used to produce 1 mole of $\text{Zn}_{1-x}\text{Cu}_x\text{O}$. Each of the mixture was prepared to produce a 20g batch. $\text{Zn}_{1-x}\text{Cu}_x\text{O}$ ceramic compound was prepared by mixing the copper oxide (CuO) and zinc oxide (ZnO). These oxides were mixed according to their molecular weight and the required amount of the samples was grind with a few drops of ethanol by using an agate motor for 1 h. And then, the required amount of the samples was grind adding a few drops of ethanol in the breaker. After that, the mixture powder was heated onto the hot plate with 100°C by stirring a glass rod for 1 h and then cooled at the room temperature, to obtain the homogeneous powder was used by ball milling machine. The mixture was first-sintered at 500°C for 1 h in an electrical furnace with increasing temperature of 20°C per minute, and then cooled to room temperature with the same rate.

Pellet Sample Preparation

The mixture powder was grinding with an agate motor for 1 h. The powder was pressed into pellet by hydraulic press at a pressure of $\sim 70\text{MPa}$. The second sintering was performed at 600°C for 1 h with the same heating rate of 20°C per minute. The pellet was polished for further characterizations. The phase formation and electrical property of second-sintered pellet sample were checked by using X-ray diffractometer and electrical resistivity and conductivity measurement. Process of the sample preparation has shown in figure 1.

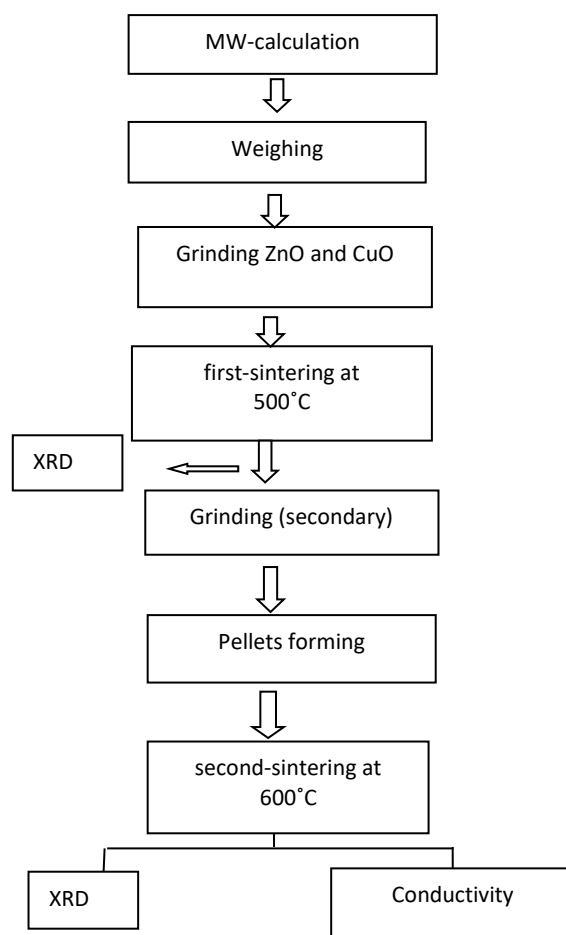


Figure.1. Process of the sample preparation

Experimental Results of XRD

Copper zinc oxide, $Zn_{1-x}Cu_xO$, has been prepared by solid state method. The phase formation of the first-sintered sample was checked by using X-ray diffractometer. The XRD spectrum of the first-sintered sample is shown in figure 2. According to the XRD characterizations of the first-sintered sample, it is confirmed that sample has been successfully formed typical hexagonal sample structure according to (ICDD card 65-3411 and 80-1916). The average crystallite size and average lattice parameter in table 1 and table 2. The average crystallite size is form to be 60.2102 nm and average lattice parameter is $a=3.3534\text{\AA}$ and $c=5.4324\text{\AA}$. The phase formation of the second-sintered sample has been checked by using X-ray diffractometer. The XRD spectrum of the second -sintered sample is shown in figure 3. According to the XRD characterizations of the second -sintered sample, it is confirmed that sample has been successfully formed hexagonal structure according to (ICDD card 89-7102 and 80-1916). The average crystallite size and average lattice parameter in table 3 and table 4. The average crystallite size is form to be 57.5455 nm and average lattice parameter is $a=3.2692\text{\AA}$ and $c=5.1749\text{\AA}$.

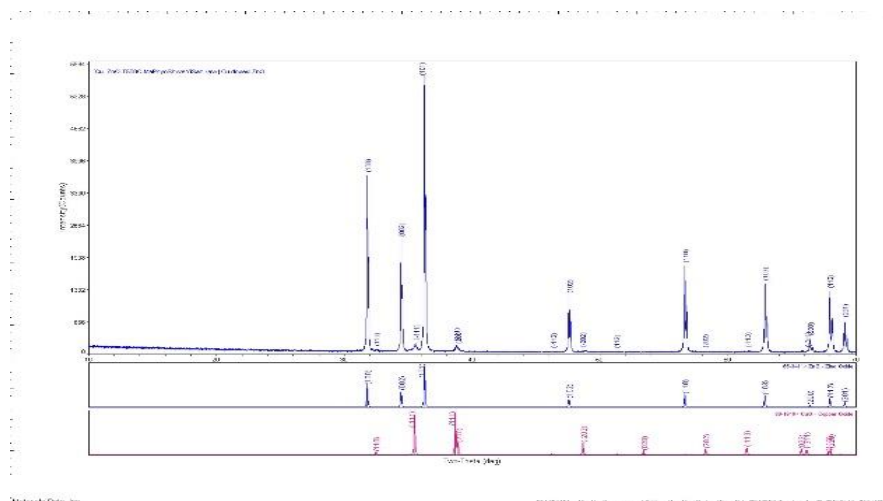


Figure.2. XRD diffractogram for first- sintered $Zn_{1-x}Cu_xO$ powder

Table.1 Crystallite size of each planes of $Zn_{1-x}Cu_xO$ powder

Plane	FWHM ~B (deg)	Peak width B(rad)	2θ(deg)	cos θ	Size D (nm)
100	0.114	0.00198	31.792	0.9618	72.4556
101	0.151	0.00263	36.274	0.9503	55.3604
110	0.233	0.00406	32.471	0.9601	35.5109
002	0.111	0.00193	34.444	0.9552	74.9277
102	0.103	0.00179	47.566	0.9151	84.2845
200	0.525	0.00916	38.853	0.9431	16.0451
112	0.229	0.00399	51.258	0.9016	38.4758
103	0.114	0.00198	62.881	0.8532	81.6765
201	0.116	0.00202	69.108	0.8236	83.1522
Average crystallite size					60.2102

Table.2 Lattice parameter of each planes of $\text{Zn}_{1-x}\text{Cu}_x\text{O}$ powder

Line	Interplanar spacing d(Å)	hkl-fcc	a(Å)	c(Å)
1	2.8123	100	3.2474	-
2	2.4745	101	3.3600	5.7928
3	2.6016	002	-	5.2032
4	1.9101	102	3.3672	5.2235
5	1.4767	103	3.5366	5.3786
6	1.3581	201	3.2559	5.5641
Average Lattice parameter			3.3534	5.4324

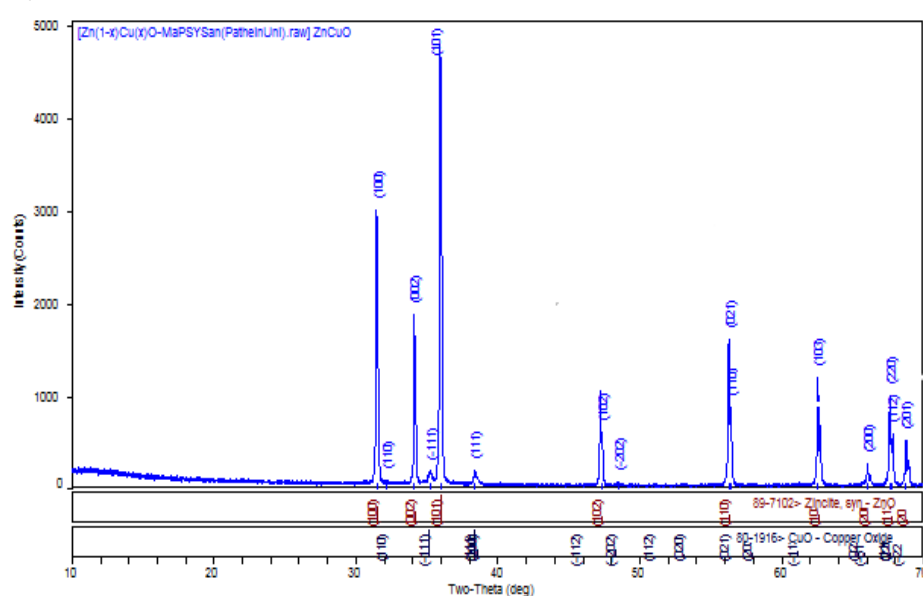
**Figure.3.** XRD diffractogram for second- sintered $\text{Zn}_{1-x}\text{Cu}_x\text{O}$ pellet

Table.3 Crystallite size of each planes of Zn_{1-x}Cu_xO pellet

Plane	FWHM ~ B (deg)	Peak width B(rad)	2 θ (deg)	cos θ	Size D (nm)
100	0.155	0.0027053	31.466	0.9625	53.247
101	0.156	0.0027227	35.949	0.9512	53.5358
110	0.093	0.0016232	32.175	0.9608	88.9017
002	0.135	0.0023562	17.0615	0.9559	61.5540
102	0.133	0.0023231	47.311	0.9159	65.2092
200	0.202	0.0035256	66.039	0.8385	46.9026
112	0.311	0.0054279	67.796	0.7783	32.8213
103	0.144	0.0025133	62.535	0.8548	64.5417
201	0.188	0.0032812	68.749	0.8254	51.1965
Average crystallite size					57.5455

Table.4 Lattice parameter of each planes of Zn_{1-x} Cu_xO pellet

Line	Interplanar spacing d(Å)	hkl-fcc	a(Å)	c(Å)
1	2.8407	100	3.2802	-
2	2.4961	101	3.2805	5.2486
3	2.6254	002	-	5.2508
4	1.9198	102	3.2673	5.2146
5	1.3811	112	3.2534	5.1367
6	1.4841	103	3.2699	5.2239
7	1.3643	201	3.2637	4.9752
Average Lattice parameter			3.2692	5.1749

Electrical Conductivity Measurements

At first, the dimensions (thickness and diameter) of the sintered pellet sample were measured by Vernier-Caliper. The dimensions of the sample were 1.55cm in diameter and 0.45cm in thickness. Temperature dependent resistances of sample were observed in the temperature range of 533K-633K. Photograph showing the experimental set up the temperature dependent electrical conductivity measurement is shown in figure 4 clearly shows that temperature dependence of the dc conductivity increases with increasing temperature. Temperature dependent resistances of the sample were measured by using Lutron LCR-9073 digital meter. K-type thermocouple was used as the temperature sensor throughout the measurement. The copper block holder was heated by 1000W heater with 500°C.

The electrical conductivities of the sample have been calculated by using formula $\sigma = \frac{l}{RA}$.



Figure.4. Temperature dependent electrical conductivity measurement

Results of Electrical Conductivity

In the present work the variation of electrical conductivity of the copper zinc oxide, $\text{Zn}_{1-x}\text{Cu}_x\text{O}$ pellet sample in the temperature range 533K-633K is shown in figure.5. In this curve, electrical conductivity of the sample is found to increase with increasing temperature. At 533K (starting temperature), the resistivity of the sample is $82.36 \times 10^6 \Omega\text{cm}$ (conductivity is $0.012 \times 10^{-6} \text{Scm}^{-1}$) and at 633K (end temperature), the resistivity of the sample is $0.5 \times 10^6 \Omega\text{cm}$ (conductivity is $2 \times 10^{-6} \text{Scm}^{-1}$) respectively. Figure.6 shows the temperature dependent electrical resistivity curve of copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) pellet sample. As shown in this curve, electrical resistivity of the sample is found to decrease with increasing temperature. Two points (data) of the sample are presented in figure.6. It indicates the high resistivity of the sample.

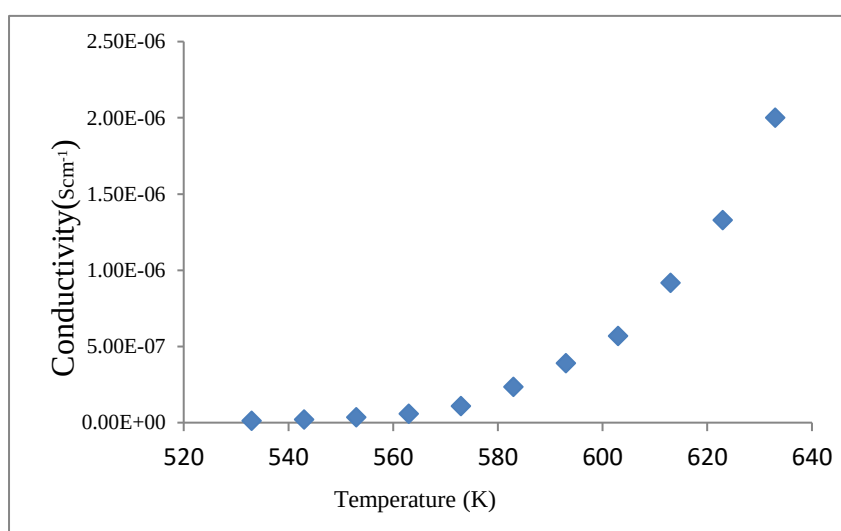


Figure.5. Plot of temperature dependent electrical conductivity versus temperature curve of ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) pellet

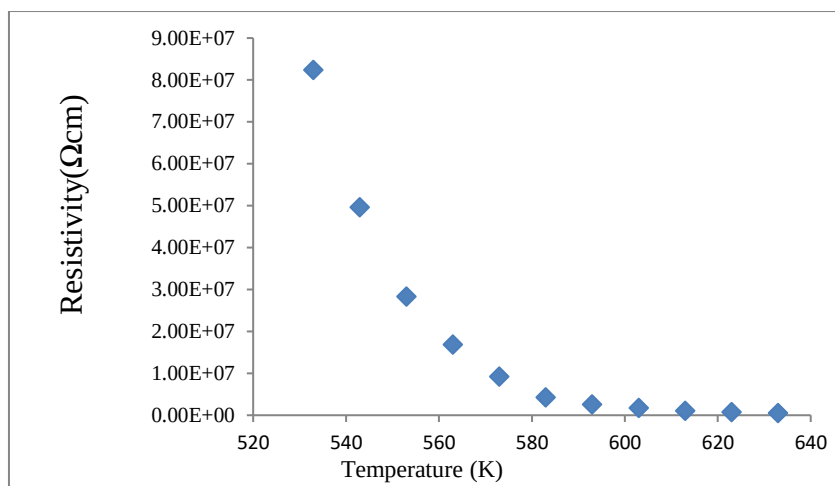


Figure.6. Plot of temperature dependent electrical resistivity versus temperature curve of $(\text{Zn}_{1-x}\text{Cu}_x\text{O})$ pellet

Discussion

Copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) has been prepared by solid state method in this paper. The XRD characterizations, the samples have been successfully formed typical hexagonal structure after the counter diffusion reaction of the metal oxides at first- sintering temperature of 500°C and second-sintering temperature of 600°C. The average lattice parameter and average crystallite size have been calculated for further characterization.

Copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) powder can be presented of average lattice parameters and average crystallite size by $a = 3.3543\text{\AA}$, $c = 5.4324\text{\AA}$ and $D = 60.2102\text{ nm}$. Copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) pellet can be presented of average lattice parameters and average crystallite size by $a = 3.2692\text{\AA}$, $c = 5.1749\text{\AA}$ and $D = 57.5455\text{ nm}$. By using XRD results were obtained hkl 100, 002, 101, 102, 021, 110, 103, 220.

Electrical conductivities of the copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) pellet sample have been received from measurement of resistances of the pellet sample. The electrical conductivity of the sample is found to increase with increasing temperature. At 533K (starting temperature), the resistivity of the sample is $82.36 \times 10^6 \Omega\text{cm}$ (conductivity is $0.012 \times 10^{-6} \text{ Scm}^{-1}$) and at 633K (end temperature), the resistivity of the sample is $0.5 \times 10^6 \Omega\text{cm}$ (conductivity is $2 \times 10^{-6} \text{ Scm}^{-1}$) respectively.

Conclusion

The main objective of this paper is to prepare and fabricate copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) powder and pellet by using solid state method. In theory, for an ideal hexagonal structure, $a = b \neq c$. According to their XRD results and calculations, the crystal structure of copper zinc oxide ($\text{Zn}_{1-x}\text{Cu}_x\text{O}$) compound can be concluded hexagonal structure. And then, the nano-size crystal structure has been investigated. The electrical conductivity of the sample is found to increase with increasing temperature, whereas resistivity decreases. Generally, the electrical resistivity of the sample decreases with the increase of temperature, which shows that sample have semiconductor behavior.

Acknowledgements

I would like to thank Professor Dr Than Tun, Rector, Patheingyi University, for his valuable suggestions for this research work.

I would like to thank Professor Dr Aye Ngwe, Head of Department of Physics, Patheingyi University, for his valuable suggestions for this research work..

I wish to express my thanks to Professor Dr Than Than Swe, Department of Physics, Patheingyi University, for her kind permission to undertake this research.

I am also thankful to professor Dr Khin Myo Ma Ma Saw, Department of Physics, Patheingyi University, for her kind permission to undertake this research.

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