SYNTHESIS OF INDIUM ZINC OXIDE VIA SOL GEL METHOD AS CONDUCTIVE FILLER

This report is submitted in accordance with requirement of the University Teknikal Malaysia Melaka (UTeM) for Bachelor Degree of Manufacturing Engineering with Honors.

by

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DECLARATION

I hereby, declared that this dissertation entitled “synthesis of indium zinc oxide via sol gel method as conductive filler” is the result of my own research except as cited in references.

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Date : 26 JUNE 2019

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APPROVAL

This report is submitted to the Faculty of Manufacturing Engineering of Universiti Teknikal Malaysia Melaka as a partial fulfilment of the requirement for the degree of Bachelor of Manufacturing Engineering (Hons).
The members of the supervisory committee are as follow:

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ABSTRACT

Electromagnetic Interference (EMI) is a problem that occurs in electronic world due to increased availability of electronic devices and needs to be overcome. To solve this problem, EMI shielding has been designed using conductive or magnetic materials to block the EMI. Metals are common materials used to make EMI shielding but due to its high corrosion and assembly cost, new materials are being developed to replace metals. This project aims to synthesize indium zinc oxide (IZO) powder that can be used as filler in polymer composite to replace the use of metal as EMI shielding. The IZO powder was formed by using the sol-gel method. In this study IZO particles synthesized via sol gel method at different doping concentration and annealing temperature were characterized, and effects of indium doping and annealing temperature on electrical properties of IZO particles were studied. The concentration of indium doping was varied at 0%, 3%, 5% and 7% for the same annealing temperature of 200 °C. Another set of parameter of samples was 5% indium concentration with different annealing temperatures of 80, 200, 300 and 450°C. The IZO particles were characterized using scanning electron microscope (SEM), energy dispersive x-ray (EDX), x-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR). For the conductivity test, 4 point-probe test was used. From the result obtained, the concentration of different indium percentages does effect the conductivity of the IZO particles and the annealing temperature also affect the microstructure of IZO particle, in which the particle size became smaller when higher temperature was applied. Annealing temperature also affects the conductivity of IZO particle, in which the conductivity of IZO increase with the increase of annealing temperature.

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ABSTRAK

Gangguan Elektromagnetik (EMI) adalah masalah yang berlaku di dunia elektronik disebabkan peningkatan ketersediaan peranti elektronik dan perlu diatasi. Untuk menyelesaikan masalah ini, perisai EMI telah direka bentuk menggunakan bahan konduktif atau magnet untuk menyekat EMI. Logam adalah bahan biasa yang digunakan untuk melindungi perisai EMI tetapi disebabkan kos kakisan dan perakitan yang tinggi, bahan-bahan baru sedang dibangunkan untuk menggantikan logam. Projek ini bertujuan untuk mensintesis serbuk indium zink oksida (IZO) yang boleh digunakan sebagai pengisi komposit polimer untuk menggantikan penggunaan logam sebagai perisai EMI. Serbuk IZO dibentuk dengan menggunakan kaedah sol-gel. Dalam kajian ini, zarah-zarah IZO yang disintesis melalui kaedah sol gel pada kepekatan doping yang berbeza dan suhu penyepuhlahan dicirikan, dan kesan doping indium dan penyepuhlindapan pada sifat elektrik zarah IZO telah dikaji. Kepekatan doping indium berubah-ubah pada 0%, 3%, 5% dan 7% untuk suhu penyepuhlahan yang sama 200 ° C. Satu lagi parameter sampel adalah 5% kepekatan indium dengan suhu penyepuhlahan yang berlainan sebanyak 80, 200, 300 dan 450 ° C. Zarah IZO dicirikan dengan menggunakan mikroskop elektron pengimbasan (SEM), sinaran penyebaran tenaga (EDX), difraksi sinar-X (XRD) dan Transformasi empatier spektroskopi inframerah (FTIR). Bagi ujian kekonduksian, ujian 4 titik-probe telah digunakan. Dari hasil yang diperoleh, kepekatan peratusan indium yang berlainan memberi kesan kepada kekonduksian zarah IZO dan suhu penyepuhlindapan juga mempengaruhi struktur mikro zarah IZO, di mana saiz zarah menjadi lebih kecil apabila suhu yang lebih tinggi digunakan. Suhu penyerapan juga mempengaruhi kekonduksian zarah IZO, di mana kekonduksian IZO meningkat dengan peningkatan suhu penyepuhlindapan.

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DEDICATION

My beloved father, Wan Omar
My appreciated mother, Che Rokiah
My adored sisters and brothers
For giving me moral support, money, cooperation, encouragement and also understandings Thank You So Much & Love You All Forever

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LIST OF ABBREVIATION

EMI	-	Electromagnetic Interference
RFI	-	Radio Frequency Interference
AC	-	Alternating Current
DC	-	Direct Current
ZnO	-	Zinc Oxide
IZO	-	Indium Zinc Oxide
ITO	-	Indium Tin Oxide
SEM	-	Scanning Electron Microscopy
EDX	-	Energy Dispersive X-Ray
XRD	-	X-Ray Diffraction
FTIR	-	Fourier Transform Infrared Spectroscopy
FED	-	Field Discharge

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CHAPTER 1

INTRODUCTION

This chapter covers brief background, problem statements, and objectives to be achieved through the study, scope of the study and significance of study.

1.1 Background of study

Electromagnetic interference (EMI), likewise called radio-frequency interference (RFI) when in the radio frequency spectrum, is an unsettling influence created by an outer source that influences an electrical circuit. The unsettling influence may debase the execution of the circuit or even prevent it from working. On account of an information way, these impacts can extend from an expansion in mistake rate to an aggregate loss of the information. Changing of electrical current and voltage from men made and natural resources can also lead to EMI. EMI is a twisting on an alternate current (AC) or direct current (DC) line sent through the conductive way of a circuit. At the point when the area of an electromagnetic field is in the radio frequency go with another electronic device, the EMI intrusion happens. EMI fields can be created by immediate and powerful remote transmitter which can disturb the activity of electronic close-by.

Electromagnetic Interference can emerge from various perspectives and from various sources. The distinctive kinds of EMI can be arranged in various ways. Man-made EMI for the most part emerges from different hardware circuits, albeit some EMI can emerge from exchanging of huge flows. From natural causes, this kind of EMI can

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emerge from numerous sources, for example, astronomical clamor and also lightning and other environmental sorts of commotion all contribute. Continuous interference is an EMI for the most part emerges from a source, for example, a circuit that is emitting a continuous signal. Anyway foundation commotion, which is ceaseless might be made in various ways, either synthetic or normally happening. Impulse noise, this kind of EMI might be man-made or normally happening. Lightning, and exchanging frameworks all add to motivation commotion which is a type of EMI.

All the electronic equipment must work with a good electrical ground system to maintain a strategic distance from issues that happen as a result of EMI. Circuit design isn't sufficient to secure against over the top dimensions of electromagnetic radiation from the equipment or impacting the rigging. Electromagnetic shielding is an elective that is typically used as a piece of these cases. Electromagnetic shield can work in the two headings which are blocking surges from the equipment and blocking electromagnetic radiation that may impact the unit. EMI shielding is known to be made for the most part from metal, which is known to have great mechanical and electrical properties. Be that as it may, metal is normally powerless to erosion issue, high thickness and high assembling expense.

Due to the properties and disadvantages of metal materials as EMI shielding, new alternatives and methods are carried out to develop the EMI shielding. One of the alternative is by the use of composite as a replacement for the use of metals in making EMI shielding. In composite world, the use of natural fibres as a reinforcement and polymer as matrix in composite structure are widely used as they provide lots of benefits and comparable mechanical properties compared to the use of metals. In order to increase the conductivity of this composite, conductive filler is added in the process of making composite.

Conductive material such as Zinc Oxide (ZnO) was used as a conductive filler in the making of composite. ZnO is a material that well known for its conductivity and high transparency as proven made by previous studies. Zinc Oxide possesses a unique position among materials owing to its superior and diverse properties such as piezoelectricity, chemical stability, biocompatibility, optical transparency in the visible region, high voltage current nonlinearity (Mujdat Caglar, 2009). To increase and

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improve the conductivity of ZnO more significantly, indium was used as dopant in making the preparation of ZnO by using the sol gel method.

1.2 Problem Statement

EMI shielding materials have been made for the most part from metal, which is known to have great mechanical and electrical properties. Be that as it may, metal is ordinarily helpless to erosion issue, high thickness and high assembling expense. Electromagnetic interference (EMI) shielding is turning into a more essential issue as electronic gadgets turned out to be all the more broadly utilized in our living surroundings and additionally in mechanical and restorative fields. With the end goal to be utilized as EMI shielding, a material requires certain level of electrical conductivity. Adding conductive filler is a technique that can be used to expand the electrical conductivity of characteristic composite.

Indium zinc oxide (IZO) doping is liked to be one of the strategies to enhance the electrical conductivity of composite. IZO covering is picked in light of the fact that contrasted with regular utilized indium tin oxide (ITO). Plus, indium zinc oxide is viewed as good with polymer composite because of its low handling temperature that makes it reasonable fir low debasement temperature of fibres.

1.3 Objectives

The purpose of this research are:

a) To synthesizes indium zinc oxide (IZO) particle via sol gel method.
b) To characterize the indium zinc oxide particle prepared at different doping concentration and annealing temperature.
c) To study the effects of indium doping and annealing temperature on electrical properties of indium zinc oxide particle.

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1.4 Scope of Research

The scopes of research are:

a) The study on synthesis of Indium Zinc Oxide particle via sol gel method.
b) The study on the effect of annealing temperature on electrical and microstructure of Indium Zinc Oxide particle.
c) The study on the effect of Indium doping concentration on electrical properties at different annealing temperature.
d) The study on characteristic of elemental crystal structure and microstructure of Indium Zinc Oxide particle.

1.5 Important of Study

Nowadays the use of electronics devices are widely around the world, without EMI shielding materials the electronic devices will have the tendency to get interrupted by the EMI. Hence, the study carried to produce EMI shielding material using conductive material such IZO as filler in composite is one of the way to produce EMI shielding by lowering the cost of manufacturing and reduce the use of metals in making the EMI shielding. As known metals are materials that have erosion issues and high assembling cost that need to be replaced to a better material.

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CHAPTER 2

LITERATURE REVIEW

2.1 Zinc Oxide

Zinc Oxide is an inorganic compound that is insoluble in water. The formula for Zinc Oxide is ZnO. It exists in the form of white powder. Zinc oxide (ZnO), a wide bandgap (3.4 eV) II–VI compound semiconductor, has a stable wurtzite structure with lattice spacing a = 0.325 nm and c = 0.521 nm (Lu, 2005). Unique properties that exhibit in ZnO such as good electrical, structural and optical properties have attracted lots of researcher to carry on experiment and utilize these unique properties to improve or develop new beneficial products. Figure 2.1 shows the physical properties wurtzite of ZnO. The scientific research was pulled in towards this material because of its different surprising electrical, basic and optical properties.

Properties	Value
Lattice constants ($T = 300$ K)	
a_0	0.32469 nm
c_0	0.52069 nm
Density	5.606 g/cm^3
Melting point	2248 K
Relative dielectric constant	8.66
Gap Energy	3.4 eV, direct
Intrinsic carrier concentration	$<10^6$ cm^{-3}
Exciton binding energy	60 meV
Electron effective mass	0.24
Electron mobility ($T = 300$ K)	200 cm^2/Vs
Hole effective mass	0.59
Hole mobility ($T = 300$ K)	5–50 cm^2/Vs

Figure 2.1: Physical properties of wurtzite ZnO (Lu, 2005).

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2.1.1 Crystal Structure of Zinc Oxide

Zinc Oxide is a wide gap semiconductor, ZnO is a long known and versatile II-VI compound semiconductor with a wide band gap of 3.44 eV, and is widely used in electronics, photonics, acoustics and sensing (Sheen-Jeff Teh, 2014). For instance, the wide band gap and simple doping procedure make this material a decent decision for different optoelectronics applications. This material additionally shows piezoelectric properties which further opens the entryway for different innovative applications The greater part of the group II–VI binary compound semiconductors take shape in either cubic zinc blende or hexagonal wurtzite (Wz) structure where every anion is encompassed by four cations at the sides of a tetrahedron, and the other way around. There are three types of crystal structure shared by ZnO as wurtzite, zinc blende, and rock salt. Wurtzite symmetry exist only in stable phase of thermodynamically under the ambient conditions. The zinc-blende ZnO structure can be stabilized only by growth on cubic substrates and the rock salt (NaCl) structure may be obtained at relatively high pressures (Bakhtiar Ul Haq, 2013). Figure below shows the structure of zinc oxide.

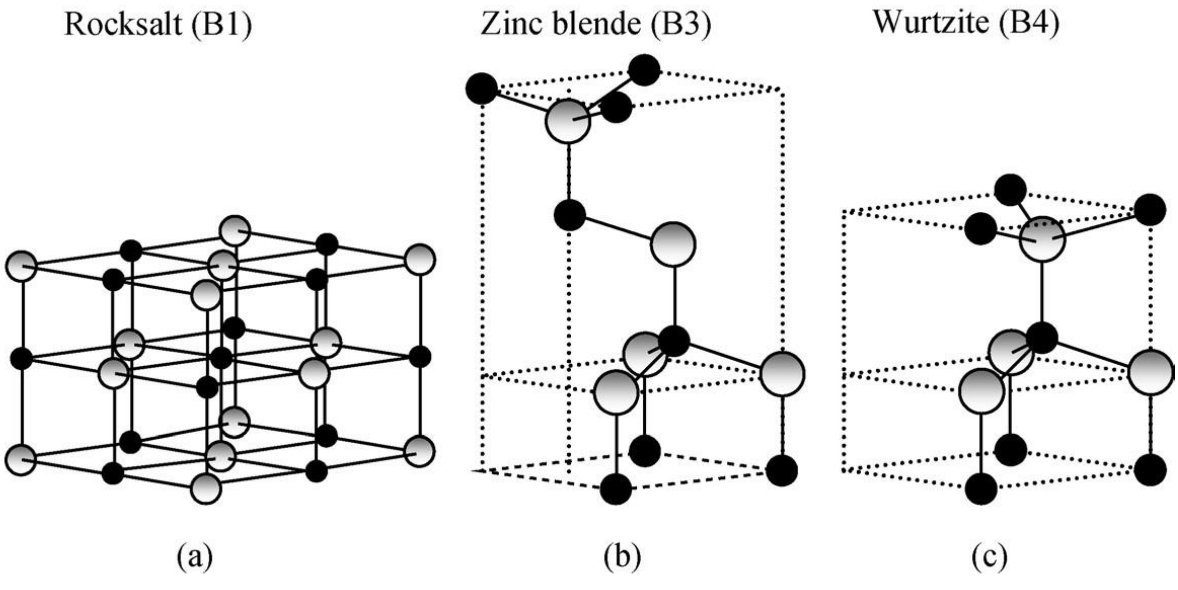

Figure 2.2: Shows (a) cubic rocksalt (B1), (b) cubic zinc blende (B3), and (c) hexagonal wurtzite (B4). Shaded grey and black spheres denote Zn and O atoms, respectively (Özgur, 2009).

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Wurtzite zinc oxide has a hexagonal structure with lattice parameters a = 0.3296 and c = 0.520 65 nm, the structure of ZnO can be simply described as a number of alternating planes composed of tetrahedrally coordinated O^{2-} and Zn^{2+} ions, stacked alternately along the c-axis (Wang, 2004). This tetrahedral coordination is common of sp3 covalent bonding nature; however, these materials additionally have a significant ionic character that will in general increment the band hole past the one anticipated from the covalent holding. ZnO is stable thermodynamically with the wurtzite phase due to its ionicity that resides exactly at the borderline between the covalent and the ionic materials (Jagadish, 2007).

2.1.2 Electrical properties of Zinc Oxide

As an immediate and wide bandgap material, ZnO is drawing in much consideration for an assortment of electronic and optoelectronic applications. Points of interest related with an enormous bandgap incorporate high-temperature and high-control activity, lower noise generation, higher breakdown voltages, and capacity to support huge electric fields. Transportation of electron in semiconductor considered to be low electric field and high electric field.

At adequately low electric fields, the vitality picked up by the electrons from the connected electric field is little contrasted with the warm vitality of electrons what's more, in this way the vitality circulation of electrons is unaffected by such a low electric field. At the point when the electric field is expanded to a point where the vitality picked up by electrons from the outer field is never again immaterial contrasted with the warm vitality of the electron, the electron circulation capacity changes altogether from its equilibrium value. The high electron mobility, high thermal conductivity, wide and direct band gap and large exciton binding energy make ZnO suitable for a wide range of devices, including transparent thin-film transistors, photodetectors, light-emitting diodes and laser diodes that operate in the blue and ultraviolet region of the spectrum (Walle, 2009).

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Table 2.1: Electrical Properties of ZnO

Electron effectiveness mass	0.28
Hole effective mass	1.8
Electron Mobility (300 K)	205 cm^{-2}
Hole mobility (300 K)	180 cm_{-2} $V_{-1}S_{-1}$
Saturation electron drift velocity	10^7 cm/S
Static dielectric constant	8.47
	1.2 C/m^2

2.1.3 N- and P- Type of doping Zinc Oxide

Despite the ongoing fast advancements, controlling the electrical conductivity of ZnO has remained a noteworthy test. While various research gatherings have revealed accomplishing p-type ZnO, there are still issues concerning the reproducibility of the outcomes and the dependability of the p-type conductivity. Indeed, even the reason for the normally watched accidental n-type conductivity in as-developed ZnO is still under discussion.

The main issues about ZnO is always about controlling the conductivity about ZnO. This is because even relatively small percentages concentrations of native point of defects and impurities (down to 10−14 cm^{-3} or 0.01 ppm) can significantly will affect the properties of electrical and as well as the optical properties of semiconductors (Walle, 2009). Hence, conductivity of ZnO can be controlled by understanding the incorporation of impurities and the role of native point defect. The unintentional n-type of conductivity of ZnO is commonly because the reason would be identified with the accidental fuse of contaminations that go about as shallow benefactors. For example, hydrogen which is available in practically all development and handling situations. By methods of density estimations, it has been appeared interstitial H shapes a solid bond with O in ZnO and goes about as a shallow giver, as opposed to the amphoteric conduct of interstitial H in conventional semiconductors.

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Acquiring p-type doping in ZnO has demonstrated to be an extremely difficult assignment. One reason is that ZnO has an inclination toward n-type conductivity, and advancement toward understanding its causes is fairly recent. ZnO has a shallow contributor level, in this way, could be the potential sources for the accidental n-type conductivity. On the other hand, these low energy intrinsic defects could be responsible for the equilibrium p-type doping difficulties of ZnO (C. H. Park, 2002). The exchange of dopants, polluting influences, and natural deformities is crucial for the electrical and optical properties of ZnO, particularly in the event that one might want to accomplish p-type conductivity. In an ongoing methodology for p-type doping by repeat temperature moodulation development it has been demonstrated exemplarily high quality crystal growth development and doping with nitrogen must be done in rather different temperature routines. Then again, particle implantation with nitrogen into industrially accessible ZnO single precious crystal and consequent thermal annealing has likewise been shown to result in the formation of a p-n junction and electroluminescence.

The creation of semiconductor devices is mainly based on ion implantation as a primary step to create p-n junctions because this method allows tailoring their depth in the material and geometrical structure in accordance with industrial production needs (G Brauer, 2011). To achieve p-conductivity, doping of ion will be considered. It has been discovered that with gathering I dopants alone, substitutional acceptors are for the most part self-compensated by interstitial benefactors, while in ZnO co-doped with H debasements, the arrangement of compensating interstitials is seriously suppressed, and along these lines the acceptor dissolvability is significantly improved by forming H-acceptor complexes. At that point, H atoms could be effectively separated from these deformity edifices at relatively low toughening temperatures, and in this way low resistivity p-type ZnO would be reachable with dopants from different group -V elements.

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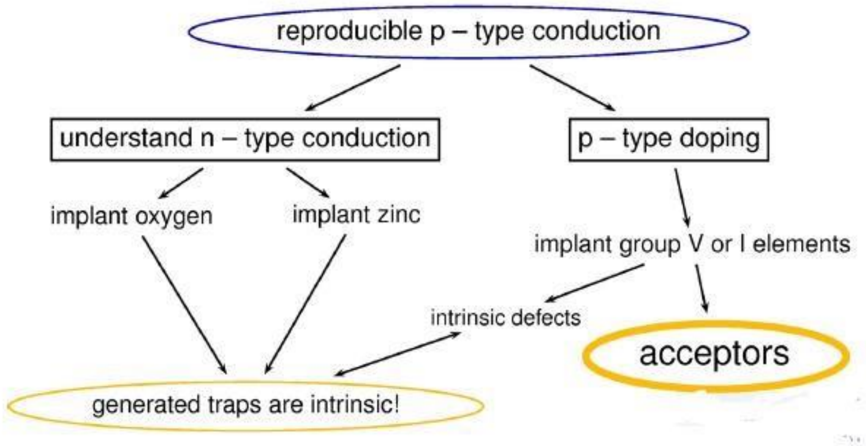

Figure 2.3: Illustration how to achieve p-type conductivity of ZnO (G Brauer, 2011).

2.1.4 Indium Zinc Oxide

Transparent conductive oxide (TCO) films such as indium tin oxide (ITO), gallium doped zinc oxide (GZO) and zinc doped indium oxide (IZO) have been studied for several applications such as solar cells, thin film transistors (TFTs) and organic light emitting diodes (OLEDs) due to their high transmittance and good electrical conductivity (G. Gonçalves, 2007). Recently, $In^{2}O^{3}$ (ZnO) system has become a particular focus of attention due to its good electro-optical properties associated with excellent chemical stability (Radhouane Bel-HadjTahar, 2014). Indium-doped zinc oxide (IZO) have been cultivated utilizing various techniques, sputtering, e-beam evaporation, pulsed laser deposition, chemical vapour deposition and including spray pyrolysis.

These amorphous IZO were accounted for to have the lesser compressive pressure and smoother surfaces than the polycrystalline ITO stored under a similar sputtering conditions. Wet etching characteristic of amorphous IZO films with enhanced electrical properties were additionally answered to be better than those of the polycrystalline ITO films since they could be etched effectively with weak acid concentration, for example, oxalic corrosive and formatted to the tapper shaped fine pitch design. In this way, the IZO is relied upon to be another possibility for transparent electrode particularly for meagre film transistor

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(TFT)- LCDs or natural ELDs. IZO composed of indium that is well known for its high conductivity and hence can improve the conductivity of the composites, compared to Indium Tin Oxide (ITO), IZO exhibit more stability (Cristina Besleaga, 2012).

In order to increase the conductivity of ZnO, materials that have good characteristic of conductivity such as Indium can be added as dopant to Zinc Oxide. Indium zinc oxide (IZO), in particular, have been attracting significant attention because of their good conductivity, high optical transparency, and low deposition temperature (Pin Zhaoa, 2018). Indium Zinc Oxide (IZO) system has demonstrated excellent optical and electrical properties, such as high optical quality, high mobility, surface uniformity and chemical and thermal stability in various environments (J.J. Ortega, 2014).

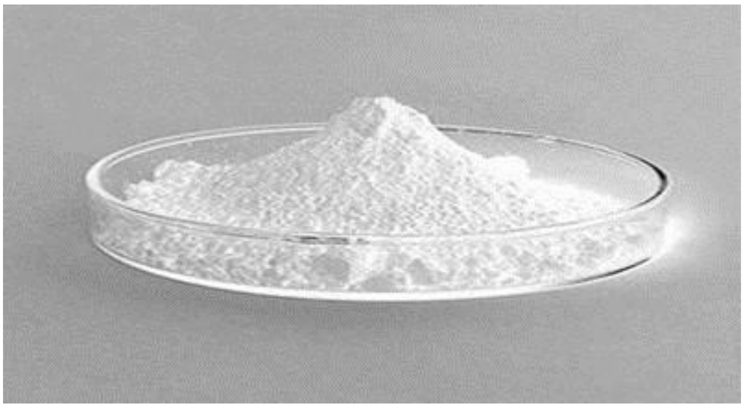

Figure 2.4: Indium Zinc Oxide Powder.

2.1.5 Sol Gel Method

Solid material can be produced from small materials by using the process of sol-gel. Sol gel method is suitable for the making of metal oxide. The typical precursor of sol gel are Zinc Alkoxides or Zinc Chloride that undergo hydrolysis and poly-condensation reactions. Hence, the sol develops towards the arrangement of a gel-like diphasic framework containing both a liquid stage and solid stage whose morphologies extend from discrete particles to constant polymer network. Sol-gel technique is widely adopted due to its comparatively simple procedure as there is no need of costly vacuum system and it has a wide-range advantage of large area deposition and uniformity of the films thickness (Ziaul

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